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FUELS TECHNOLOGY: A STATE-OF-THE-ART REVIEW

BATTELLE COLUMBUS OHIO

PREPARED FOR
NATIONAL ENVIRONMENTAL RESEARCH CENTER

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A STATE-OF-THE-ART REVIEW

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The report gives results of a state-of-the-art review of various fuel-cleaning, fuel-conversion, and emission control technologies. It includes the following classes of technologies: physical and chemical coal cleaning, residual oil desulfurization, coal refining (liquefaction), coal and oil gasification, fluidized-bed combustion of coal, and stack gas cleaning. For each technology, the report presents the extent of current practice and the status of systems under development.

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April 1975

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ABSTRACT

A state-of-the-art review of various fuel-cleaning, fuel-conversion, and emission-control technologies was conducted. Classes of technology included in the study are: physical coal cleaning, chemical coal cleaning, residual oil desulfurization, coal refining (liquefaction), coal and oil gasification, fluidized-bed combustion of coal, and stack gas cleaning. For each technology the extent of current practice and the status of systems under development is presented.

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Multiply English Unit	by Conversion	To Obtain Metric Unit
acres	0.405	hectares
acre-feet	1233.5	cubic meters
barrel, oil	158.97	liters
British Thermal Unit	0.252	kilogram-calories
British Thermal Unit/pound	0.555	kilogram calories/kilogram
cubic feet/minute	0.028	cubic meters/minute
cubic feet/second	1.7	cubic meters/minute
cubic feet	0.028	cubic meters
cubic feet	28.32	liters
cubic inches	16.39	cubic centimeters
degree Fahrenheit	0.555(°F-32)[(a)]	degree Centigrade
feet	0.3048	meters
gallon	3.785	liters
gallon/minute	0.0631	liters/second
horsepower	0.7457	kilowatts
inches	2.54	centimeters
inches of mercury	0.03342	atmospheres
pounds	0.454	kilograms
million gallons/day	3785	cubic meters/day
mile	1.609	kilometer
pound/square inch (gauge)	(0.06805 psig+1)[(a)]	atmospheres (absolute)
square feet	0.0929	square meters
square inches	6.452	square centimeters
tons (short)	0.907	metric tons (1000 kilograms)
yard	0.9144	meters

(a) Actual conversion, not a multiplier.

FINAL REPORT

on

FUELS TECHNOLOGY
A STATE-OF-THE-ART REVIEW

to

ENVIRONMENTAL PROTECTION AGENCY

from

BATTELLE
Columbus Laboratories

March 14, 1975

by

E. H. Hall, D. B. Peterson, J. F. Foster,
K. D. Kiang, and V. W. Ellzey

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INTRODUCTION

The steadily increasing demand for energy in the United States and around the world combined with the need to consider environmental factors have given impetus to a large number of developments in many energy-related technologies. This study was conducted for the Control Systems Laboratory of the U. S. Environmental Protection Agency to provide a current assessment of the status of these varied activities. The study included the following technologies.

Fuel Cleaning

1. Physical coal cleaning
2. Chemical coal cleaning
3. Residual oil desulfurization

Fuel Converstion

4. Coal refining (liquefaction)
5. Coal and oil gasification, low- and intermediate-Btu gas
6. Coal gasification, high-Btu gas

Emission Control

7. Fluidized-bed combustion
8. Stack gas scrubbing.

For each of these technologies the objective was to assess the current status of the technology in terms of: the extent of actual application, the types of research and development projects in progress, the operating histories of pilot or demonstration plants, the problems which remain to be solved, and current projections for the completion of each development stage.

The format of the report has been designed to facilitate periodic updating of the information to keep it current. Each of the technologies listed above is treated in a separate section of the report. Where appropriate, specific processes within a given technology are described briefly and a flow chart is included to show the process principles. This is followed by a state-of-the-art table which includes the following information: the type of facility; the location; the owner or contractor; the status/operating history; the name, address, and telephone number of a contact person knowledgable with respect to the process; and a list of references pertinent to the specific process. This basic information has been presented as concisely as possible so that the reader may turn to a specific process and quickly find the pertinent details regarding the status of the process. The numbering of pages, tables, and figures is distinct within each section so that additional processes can be included readily at a later date, if desired.

SUMMARY AND CONCLUSIONS

The status of each of the eight energy technolgies under consideration in this study varies from the development stage to established commercial practice. To provide an overview of the current situation, the status of each technology is summarized briefly in the following paragraphs.

Physical coal cleaning is an established commercial technology. Conventional coal washing cannot remove sulfur bound to the coal structure and is thus limited in its effectiveness. Nevertheless, it is a useful technique which represents a partial solution to the problem of SO_2 emissions from coal combustion. Experimental studies are in progress in an attempt to find improved cleaning methods. Such advanced methods all are faced with the problem of developing equipment capable of high throughput rates.

Chemical coal cleaning is in the early development stage. No pilot plants have been built to date. Chemical coal cleaning processes promise more complete removal of sulfur than physical coal cleaning at a lower cost than conversion of coal to clean liquid or gaseous fuels.

Desulfurization of residual oil is an established technology which has not been practiced widely in the United States for economic reasons. High-sulfur resid has generally been used in maritime applications or exported. This is expected to change as these outlets become inadequate to absorb the volume of high sulfur resid produced. Direct or indirect desulfurization, gasification, or fluidized-bed combustion of high-sulfur residual oils will have to be employed to utilize the heavy fractions in an environmentally acceptable manner.

Coal refining or liquefaction processes are in various stages of development. The Bergius process was used in Germany during World War II but no active development of the process is underway now. The largest scale developments of other processes include a 1.5 ton-per-hour COED pilot plant and a 2 ton-per-hour SRC pilot plant. The latter is not yet completely operational.

Coal gasification technology is both commercial and developmental. First generation systems, such as Lurgi, Wellman-Galusha, Winkler, and Koppers-Totzek, are commercially available. Lurgi units are to be used in commercial-scale SNG plants now in the planning stage. Advanced gasification systems have not been developed to demonstration scale at this time. The Hygas, 3 ton per hour pilot plant, and the CO_2 Acceptor, 1.5 ton-per-hour pilot plant represent the largest scale in this country. A 4 ton-per-hour COGAS pilot plant has been operated in England and a smaller PDU unit of the same type has been operated at Plainsboro, New Jersey.

Fluidized-bed combustion of coal is being developed in both atmospheric and pressurized systems. A 30 MW atmospheric fluidized-bed boiler is under construction in West Virginia. Pressurized systems show promise of excellent combustion efficiency and SO_2 and NO_x control. The largest unit is a 12.5 inch diameter (about 0.65 MW) miniplant at Linden, New Jersey.

Stack gas scrubbing is a demonstrated technology which has been difficult to reduce to sound engineering practice with high reliability. The number of operating units is increasing and experience gained is helping in the solution of problems. Recent installations are showing higher availability factors than early test units. The total capacity of operational units and units under construction is about 8,000 MW with an additional 26,000 MW planned.

It is evident that progress is being made across a broad front in energy technology. Some techniques for reducing the environmental impact of fuel combustion are available now and should be used as effectively as possible. Other technologies under development will provide a range of options so that the best strategy for fuel use can be adopted in each situation.

SECTION 1

PHYSICAL COAL CLEANING

Introduction

Physical coal cleaning, or mechanical cleaning, is the process of removing ash (rock) and sulfur (pyrite) from coal by a process other than chemical modification or destruction of either the coal or the impurities. Thus, physical coal cleaning is generally unable to remove impurities that are chemically combined with the coal substance. Sulfur incorporated into the chemical structure of the coal ("organic" sulfur) is a major example. Most physical cleaning processes use water and are commonly called coal washing.

The cleaning process can be based on any physical or physical-chemical difference between the coal and the impurity. The most commonly employed property is density: "pure" bituminous coal having a specific gravity of about 1.3, "rock" about 2.0 to 2.7, and pyrite about 4.8-5.0. The physical-chemical surface properties are also employed in the froth flotation process.

The impurities may be derived from the roof or floor of the mine or be imbedded in the coal seam itself. Impurities may occur as massive pieces, as microscopic particles, or in intermediate sizes. Crushing the coal physically releases the impurity particles from the coal substance.

History

Physical coal cleaning has been practiced in some form as long as coal has been mined. Originally, hand picking was used to remove large pieces of rock from the mined coal. Later, mechanical pickers, whose operation depended on differences in the shape of coal and rock (largely slate) particles were developed. Such processes became increasingly impractical with the development of mechanized mining which reduced the average size of both coal and impurity. Various "washing" processes, borrowed from the field of one beneficiation and largely based upon separation by specific gravity, have become the major coal cleaning processes.

After about 1945, the electric utilities began to dominate the coal market, and pulverized coal firing became almost exclusively the method of firing utility boilers with coal. Under these circumstances, the conclusion was reached that the most economical way of separating the coal, ash, and sulfur was by burning in the utility boiler furnace, and the practice of coal cleaning diminished.

Table 1.1 shows the recent trend in coal cleaning, characterized by a steady decline in both the total annual tonnage cleaned and the percentage of the total annual production cleaned. This trend was interrupted in 1972.

Reasons for Coal Cleaning

Coal was originally cleaned to improve its appearance to the customer. However, the removal of ash minerals increased the heating value, reduced the ash problems encountered by users, and reduced shipping costs per unit of heating value.

Current interest in coal cleaning for steam coals centers on sulfur reduction. Various state and federal regulations have been or are expected to be applied to limit the emissions of sulfur oxides. These regulations effectively would limit the sulfur content of coal to about 1 percent. The supply of such coals in the Eastern United States is quite limited, and a method of economically reducing the sulfur content of eastern coals could have great value. Potential future interest in coal cleaning may result from restrictions on the release of lead, cadmium, mercury, etc, from coal burning equipment. These potential pollutants are removed in part by the cleaning process.

It is not possible to clean eastern coals as a class to 1 percent sulfur or less. For this reason, attention has been diverted to other methods of complying with sulfur emission regulations, such as stack gas scrubbing, gasification, and chemical refining. However, coal cleaning

TABLE 1.1. PRODUCTION AND CLEANING OF BITUMINOUS COAL AND LIGNITE IN THE UNITED STATES (U.S. DEPT OF INTERIOR BUREAU OF MINES MINERALS YEARBOOK)

	10^6 tons								
	1964	1965	1966	1967	1968	1969	1970	1971	1972
Total Production	487	512	534	553	545	561	603	552	595
Quantity Cleaned	310	332	341	349	341	335	323	271	293
Percentage Cleaned	63.7	64.9	63.8	63.2	62.5	59.7	53.6	49.1	49.2

offers a known route to a significant reduction in sulfur emissions, and may be of assistance in connection with other clean-up processes. For example, a reduced sulfur content in the coal would permit use of a less efficient stack gas scrubber. Also, a reduced sulfur content would reduce the hydrogen consumption of some chemical coal refining processes.

While the total impact of universal coal cleaning has not been evaluated, a recent Bureau of Mines publication(1)* permits a rough estimate of the impact. Based upon samples from 322 mines, mostly in the eastern United States, the average sulfur content of the coal could be reduced from 3.2 percent to 2.3 percent at a 90 percent yield when crushed to a top size of 1-1/2 inch. Similarly, coal crushed to 3/8 inch and 14 mesh would yield sulfur levels of 2.0 and 1.8 percent, respectively. Simultaneously the ash content would be reduced from an average of 14 percent to 7.2 percent, 6.5 percent, and 6.0 percent, at the same levels of coal size. Slight additional gains could be obtained by cleaning at lower yields.

State of the Art

Table 1.2 lists the types of equipment and processes now dominating the coal cleaning field. A typical coal cleaning plant will use several types of equipment or processes. Figure 1.1 is a flow diagram for a modern coal cleaning plant. Some plants may replace the dense media vessel by jigs, the dense media cyclones by tables,etc. Coal cleaning plants may also use series cleaning, where the product or waste stream from one process is sectioned by another process.

Current Research

Research in coal cleaning on related topics is carried out by the U.S. Bureau of Mines, the manufacturers of coal cleaning equipment, and various universities. The research done by manufacturers of coal cleaning equipment is regarded as proprietary and little information is available concerning such research. It is believed to be largely concerned with optimizing equipment and plant design. The Bureau of Mines, with EPA sponsorship has a research program ranging from optimization of existing equipment to explorations of new cleaning concepts.

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*References for this Section are given on Page 1-9.

TABLE 1.2. TYPES OF EQUIPMENT USED IN CLEANING BITUMINOUS COAL AND LIGNITE IN THE UNITED STATES (U.S. DEPT. INTERIOR BUREAU OF MINES MINERALS YEARBOOK)

	10^6 tons cleaned*					
	1967	1968	1969	1970	1971	1972
Jigs	161	159	155	140	115	128
Tables	50	47	45	44	36	40
Classifiers	4	5	3	4	2	3
Launders	5	4	5	5	5	5
	219	215	208	193	158	176
Dense Media						
Magnetite	66	71	72	77	70	75
Sand	33	27	24	23	18	15
$CaCl_2$	3	2	2	2	2	2
	101	99	98	102	90	92
Flotation	8	9	10	11	9	13
Pneumatic	21	17	19	18	15	12
Total	349	340	335	323	271	293

*Rounding errors not adjusted.

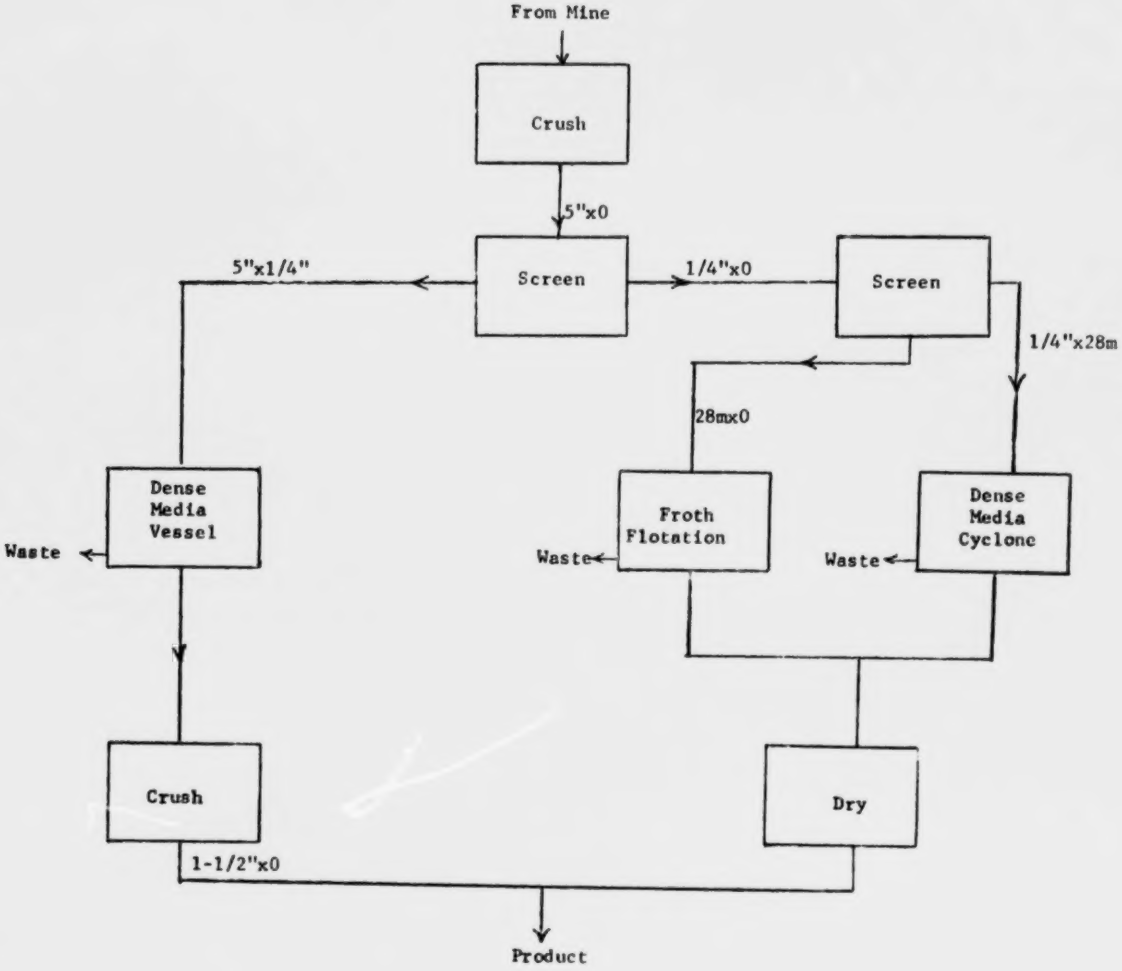

FIGURE 1.1. FLOW DIAGRAM FOR A TYPICAL MODERN COAL CLEANING PLANT

Overall efficiency, 85 to 90 percent.

The difficulty with most of the new* cleaning concepts is in devising equipment that can economically treat the large tonnages involved in coal cleaning.

Table 1.3 lists the recent and current research in new coal cleaning processes. A short description of these processes is given in Appendix 1A,which follows this Section. It may be noticed that these new processes are generally directed towards the cleaning of fine coal, about 28 mesh and smaller. Particle dynamic effects make compact, high-capacity equipment for cleaning fine coal particularly difficult to achieve.

Concluding Remarks

Several features make coal cleaning a difficult field for significant technical inovation. First, the field itself is old and has been extensively developed. Second, most of the coal cleaning processes are adaptations from the field of ore beneficiation which has perhaps justified a higher level of research and development than coal itself. Third, the existing methods, when properly applied, do an excellent job of separating coal from mineral impurities when they exist as separate particles, and no physical process can separate the coal and impurity combined in a single particle. Thus, a new process can offer little or nothing in terms of its separating ability. To improve separations, coal must be cleaned in finer sizes, and the capacity of a cleaning device is generally inversely proportional to the particle size of the coal being cleaned. For this reason, most coal cleaning plants do not fine crush or grind before cleaning, even if the cleaned coal will subsequently be crushed at the plant (see Figure 1.1).

If a significant technical innovation should occur in coal cleaning, it will almost certainly be in the cleaning of fine coal. However, the existing froth flotation processes make a good separation on fine coal and hence represents an entrenched competition to any new process. Its disadvantage is in producing a wet product which normally must be dried before shipment.

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*Most, but not all, of the "new"coal cleaning concepts are attempts to adapt mineral beneficiation processes developed in other fields.

TABLE 1.3. STATUS OF NEW COAL CLEANING PROCESSES

Process	Organization	Status	Reference	Remarks
Two Stage Froth Flotation	Bureau of Mines	Entering Fullscale (12 ton/hr) Prototype	2,3	Considered promising
Differential Grinding and Classification	Bureau of Mines	Laboratory	4	
	Bituminous Coal Research, Inc.	Pilot Plant	5	Currently inactive
	ILOK	Unknown	6	
Electrostatic		Laboratory	4,7	
Electrophoretic	Bureau of Mines	Laboratory	8	
High Gradient Magnetic Separation	MIT	Laboratory	3	
"Chemical" Comminution	Syracuse University Research Corp.	Laboratory	9	

References for Section 1

(1) Deurbrouck, A. W., "Sulfur Reduction Potential of the Coals of the United States", U.S. Dept. of Interior, Bureau of Mines Report of Investigations 7633 (1972).

(2) Miller, K. J., and Baker, A. F., "Flotation of Pyrite from Coal", U.S. Dept of Interior Bureau of Mines Technical Progress Report 51 (Feb. 1972).
Miller, K. J., "Flotation of Pyrite from Coal: Pilot Plant Study", U.S. Dept. Interior Bureau of Mines Report of Investigations 7822 (1973).

(3) Private communication from A. W. Deurbrouck, Bureau of Mines.

(4) Abel, W. T., et al, "Removing Pyrite from Coal by Dry-Separation Methods", U.S. Dept of Interior Bureau of Mines, Report of Investigations 7732 (1973).

(5) Saltsman, R. D., "The Removal of Pyrite from Coal", ASME Publication 68-WA/FU-2 (December 1968).

(6) Foster, J. F., et al, "Assessment of the Potential for Collodial Fuels in Department of Defense Applications", Battelle Columbus Laboratories report to Defense Advanced Research Projects Agency, Contract No. DAAH01-74-C-0837, AO No. 2758 (August 15, 1974).

(7) Glenn, R. A., and Grace, R. J., "A Study of Ultrafine Coal Pulverization and Its Application", U.S. Dept. of Interior, Office of Coal Research, Research and Development Report No. 5 (1963).

(8) Miller, K. J., and Baker, A. F., "Electrophoretic - Specific Gravity Separation of Pyrite from Coal", U.S. Dept. of Interior Bureau of Mines, Report of Investigations 7440 (October 1970).

(9) "Chemical Comminution Shows Promise for Coal", Chemical and Engineering News, pages 16-17 (Septebmer 2, 1974).

Appendix 1A

Description of New Coal Cleaning Processes

Two-stage Froth Flotation. Conventional froth-flotation coal cleaning uses frothing agents and conditions which float the coal from the mineral matter. This results in particles containing both coal and pyrite, and very fine pyrite particles, appearing in the cleaned coal. A second froth flotation process, which floats the pyrite from the coal, removes these mixed and fine particles and yields a cleaner product.

It is reported that negotiations are underway to install a full scale prototype in an existing coal cleaning plant. The fine coal stream is 12 tons/hr.

Differential Grinding and Classification. The Bradford breaker, dating to 1874, is an early example of this process. Typically, the impurities in coal are more resistant to crushing or grinding than is the coal. Thus, after crushing or grinding the impurity particles are larger than the coal particles and can be separated by some classification step, in current concepts, by an air classifier after a fine grinding step. Although moderately successful technically, the required equipment appears to be too large and expensive to justify the moderate sulfur reduction obtained, even when carried out at the utility plant where the coal is ground for pulverized coal firing.

Electrostatic Cleaning. A number of laboratories have investigated separating coal and mineral matter by employing differences in resistivity and dielectric constant. Generally restricted to treating fine coal, the processes work but a machine with economical capacity has not been devised.

Electrophoretic Cleaning. This process is based on the differential motion of coal and mineral matter in a suspending fluid when subjected to an electric field. The process effects a separation, but economical machinery has not been envisioned.

High Gradient Magnetic Separation. This process, which depends on a high gradient, in contrast to a high field, is one of the newer separation processes. Little information is available concerning its application to coal cleaning. It will probably work in the technical sense, but devising a machine of realistic capacity will probably be difficult.

"Chemical" Comminution. This process is akin to differential grinding. A liquid agent, such as methanol or ammonia, causes the coal to fragment along existing fracture planes without affecting the mineral matter. The fragmented coal and unaffected mineral matter can then be separated by some conventional cleaning process.

SECTION 2

CHEMICAL COAL CLEANING

Introduction

The processes for chemical cleaning of coal expose the mined coal to chemicals which dissolve or convert undesirable impurities preferentially into forms that can be easily separated. The treatment is primarily directed at the removal of sulfur compounds that otherwise would appear as gaseous waste products when the coal is burned or converted to liquid and gas fuels. Coal can also be chemically treated to remove ash, but the low-ash product may not justify the cost of the extra step. Washed or mechanically cleaned coal may be a preferred feed to the chemical cleaning process to reduce the chemicals consumed and the costs for cleaning high-ash coals.

Chemical cleaning is most effective in dissolving the discrete particles of iron sulfide mineral occluded among the layers of the coal seam. Coal also contains varying proportions of sulfur combined as sulfur compounds in the nonmineral structure. This organic sulfur is highly resistant to chemical treatment and the undissolved part is discharged as sulfur-containing gases when the coal is used. Thus, the choice between sulfur removal by pretreatment with chemicals or by post combustion flue gas cleaning (Section 8) depends upon the particular coal to be treated and the level of sulfur that can be tolerated in the products of conversion or combustion.

This section describes individually the

TRW-Meyers process,
Battelle process,

neither of which has advanced to as large-scale testing as have the flue-gas cleaning processes described in Section 8. The chemical cleaning processes differ primarily in the chemicals employed and in the results obtained. The TRW-Meyers process removes only the pyritic surfur from the coal, but the Battelle process is said to remove the pyritic sulfur and a significant portion of the organic sulfur.

2.1 Chemical Cleaning Processes

2.1.1 TRW-Meyers Process

This process employes a chemical leach of the coal with aqueous ferric sulfate at temperatures of between 50 and 130 C. The reagent is selective for pyrite, removing 83-98 percent of the pyrite in 19 different coals tested. The reduction in total sulfur was 40 to 82 percent. The products of the reaction are dissolved ferrous sulfate and sulfuric acid, and precipitated free sulfur. The depleted leach solution is removed by drainage and rinsing after which it is regenerated to ferric sulfate by oxidation with air. Precipitated free sulfur is washed out with an appropriate solvent or vaporized and recovered by condensation.

TABLE 2.1.1. TRW-MEYERS PROCESS FOR CHEMICALLY CLEANING

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Bench Scale	Redondo Beach, Calif.	TRW, Inc. (EPA support)	1971-present. Continuous leaching of crushed coals with ferric sulfate solution to determine leaching characteristics and recoveries. Obtained data for pilot plant design. Capacity about 2 lb per hr.
Pilot Plant	Same	Same	1973. Design study for pilot plant completed. Capacity, 500-1000 lb coal/hr Sept. 1974. Contract for construction and operation of pilot plant being negotiated.
	(Contact: R. A. Meyers, Applied Technology Division, TRW Systems Group, Redondo Beach, California, September 24, 1974 Tel. 213-535-1549		

Reference

J. W. Hamersma, M. L. Kraft, W. P. Kendrick and R. A. Meyers, "Chemical Desulfurization of Coal to Meet Pollution Standards", American Chemical Society, Divison of Fuel Chemistry Preprints Volume 19, No. 2, pp 33-42. Paper presented at 167th National Meeting, Los Angeles April 1-5, 1974. Also an adaptation of the paper in Coal Mining and Processing, Volume 11, No. 8, pp 36-39 (August 1974).

2.2.2 Battelle Process

This is a proprietary process concerning which no details are to be released until its patent position has been established. It is stated that "The feasibility of producing low sulfur coal by chemical desulfurization has been established in laboratory scale experiments. Heating a variety of coals in aqueous solutions at elevated temperatures and pressures extracts the pyritic sulfur and the sulfate sulfur along with a significant portion of the organic sulfur."

SECTION 3

DESULFURIZATION OF RESIDUAL FUEL OIL

Introduction

At the present time, only .07 percent of U.S. residual fuel oil capacity is being desulfurized in the United States because of the prohibitive economics inherent in the processes known for such desulfurization. Some low sulfur residual fuel oil has been produced and used domestically, but this residual product has been derived from the refining of very sweet ($\leq$0.15 percent S) crude oil from either domestic or foreign sources.

The demand for residual fuel oil in the United States has been increasing at a rate of from 4 to 7 percent per year over the past few years, and is expected to continue. In 1972 the supply of the product was 925.6 million barrels, made up from U.S. producers, imports, and inventory changes(1)*. Of this 1972 supply, a total of 591.7 million barrels of residual fuel oil were imported into the United States, while the United States produced 292.5 million barrels. Imports of desulfurized residual fuel oil have come mainly from the Caribbean, with small amounts from Canada's Shaheen's Isomox Plant, Italy, and Venezuela. The countries in the Caribbean supplying low-sulfur residual fuel oil are the Bahamas, the Netherland Antilles, and Trinidad. These countries have been able to desulfurize residual fuel oil commercially at a profit because of proximity to large producing fields, availability of natural gas for plant fuel, circumvention of certain U.S. import quotas, low taxes or tax exemptions, the availability of deep water unloading sites, and less stringent environmental requirements than in the United States.

The sulfur contained in the crude oil tends to be concentrated in the heavier fractions during the refining operations. When refineries are operated to maximize the yield of lighter fractions, such as gasoline, aviation fuel, and distillate oil, the yield of residual fuel oil is relatively low and the sulfur content is high even if the original crude had a low sulfur content. The common practice is to sell such high-sulfur residual oil to the maritime industry or to export it.

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* References cited in this section are given on page 3-6.

Process Description for Residual Oil Desulfurization

Two approaches may be employed for the desulfurization of residual oils. Indirect desulfurization consists of vacuum distillation, hydrodesulfurization of the vacuum gas oil, and reblending with a small portion of the bottom fraction. Most of the foreign and domestic low-sulfur-fuel-oil capacity is based on such indirect processing. This approach is not entirely satisfactory from the standpoint of supplying the demand for low-sulfur fuels as none of the heavy, high-sulfur vacuum bottom fraction is processed. In direct desulfurization the entire residuum is processed to yield low sulfur fuel oil. This is basically accomplished by a catalytic hydrogen treatment. The difficulty in this type of approach lies in the wide variation in feedstock properties when processing different heavy fractions from various original crude oils.

Specific processes which could be used are generally proprietary; some details of the H-Oil process were recently published(2,3). The H-Oil process employs an ebulliated bed of catalyst. The heavy oil and hydrogen are passed upward through an isothermal reactor (700-800 F) containing the catalyst. The desulfurization follows a psuedo second-order rate equation since the reaction is proportional to the square of the concentration. To solve the problems of reduced desulfurization rate as the sulfur concentration is reduced, the process uses a series of staged reactors. Another problem is catalyst aging. High catalyst efficiency is obtained by using a catalyst counterflow. Fresh catalyst is introduced into the last stage and spent catalyst is withdrawn from the first stage. It is reported that the H-Oil process can be made flexible with respect to feedstock characteristics and product line demand. A flow diagram of a multipurpose plant is shown in Figure 3.1. The required hydrogen is produced from light ends and no hydrocarbon raw material is required other than the

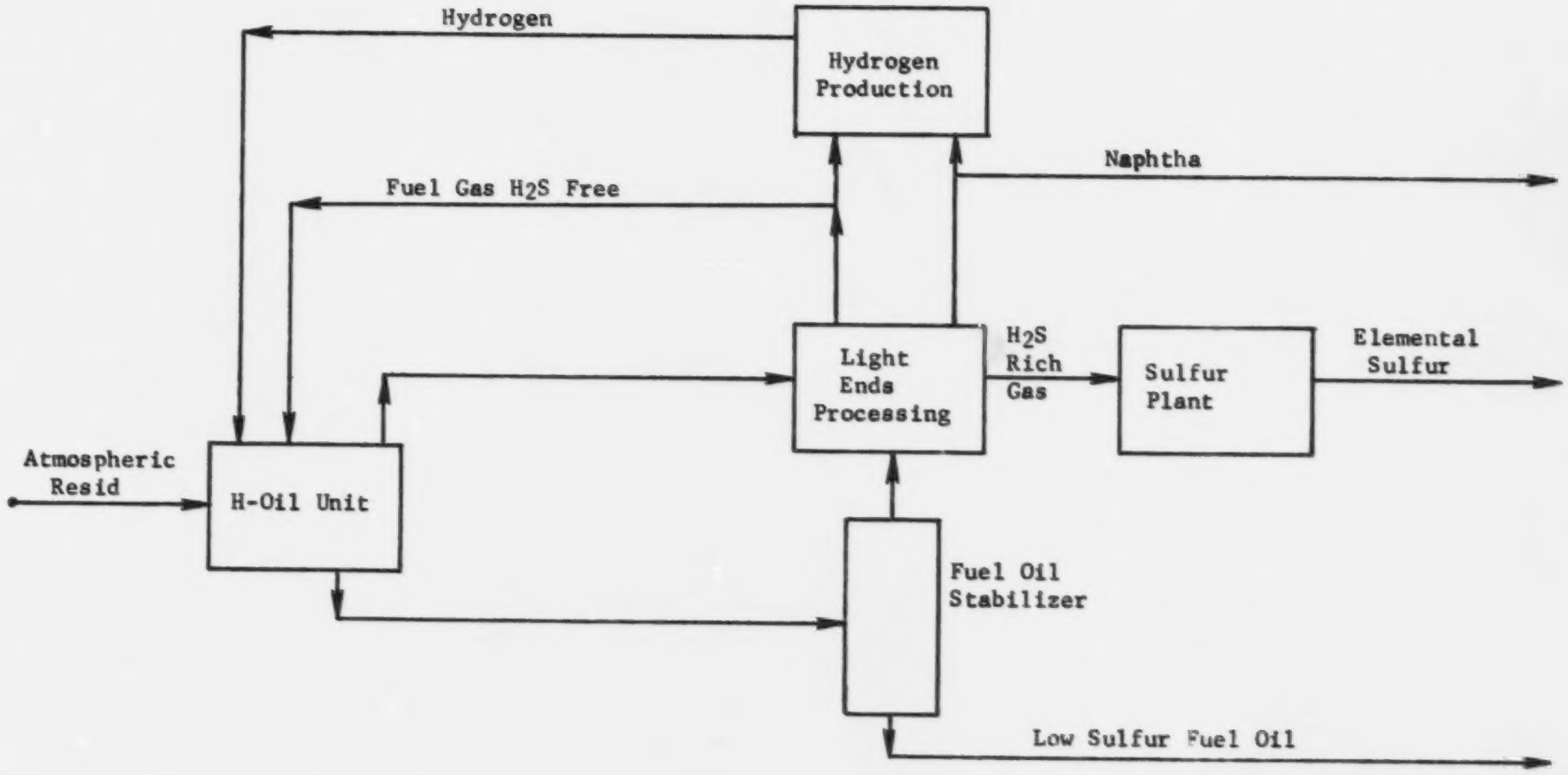

FIGURE 3.1. BLOCK FLOW DIAGRAM FOR MULTI-PURPOSE DESULFURIZATION PLANT
(Source: Reference 3)

fuel oil feed. The process has been in operation at the Lake Charles, Louisiana refinery of the Cities Service Oil Company since 1967. The capacity is now 6,000 bbl/day. Total process costs were reported to range from 88 to 138 cents per barrel of feed depending upon the type of feed. The ratio of fuel oil product to feedstock charge ranged from 0.94 to 0.98.

Foreign Plans for Desulfurization of Residual Fuel Oil

Due to the expected increasing international demand for low-sulfur residual fuel oil, there are many plans by both American and foreign companies to expand production capacity, but practically all outside this country. Some of these plans,(4) announced or underway are as follows:

- By the end of 1973, Shaheen Natural Resources, a Canadian firm, planned a new refinery for Newfoundland and Refinery, Ltd. The throughput of the refinery is 100,000 bbl/day, and it has been estimated that some 35,000 bbl/day of low-sulfur residual oil will be produced.
- By the end of 1973, Creole, a subsidiary of Exxon, planned to expand its capacity by 50,000 bbl/day for low-sulfur residual fuel oil.
- Another desulfurization plant in the Bahamas was planned for startup in 1973, by the Bahamas Oil and Refinery Company, owned by New England Petroleum Company and Standard Oil and Refining Company. It had planned to expand its crude capacity from 250,000 to 400,000 bbl/day.
- In 1973, Ameranda-Hess, in the Virgin Islands planned to expand its existing facility for production of low-sulfur residual oil by an unknown quantity.

- By 1975, a new Virgin Islands hydrodesulfurization plant will come on stream. The Virgin Islands Refining Company will produce low-sulfur residual oil by an indirect process; this plant is currently under construction by Procon, a subsidiary of UOP.
- Shaheen Natural Resources plans other desulfurization plants. A $223 million refinery is to be built in Nova Scotia at Park Haekensberry on the Strait of Conso. Plans include a 200,000 bbl/day refinery, from which 80,000 bbl/day of low-sulfur residual fuel oil would be produced.
- There is also conjecture on other desulfurization plants. For example, Shell is building a new plant in Curacao, but it is unknown whether it is an additional refining facility or a replacement for an existing facility.

Residual Oil Desulfurization Capacity and Production in the United States

The following is a tabulation of desulfurization capacity in the United States as of January 1, 1974, (no statistics could be found concerning actual production):(5)

CAPACITIES OF UNITED STATES FACILITIES FOR DESULFURIZATION OF RESIDUAL FUEL OIL

(Charge capacity in barrels per stream day)

State	Refinery	Capacity
Kansas	CRA Incorporated, Phillipsburg	4,500
Louisiana	Cities Service Oil Company, Lake Charles	6,000
Total		10,500

References for Section 3

(1) Preprint from the 1972 Minerals Yearbook, U.S. Bureau of Mines, U.S. Department of the Interior.

(2) Gregoli, A. A., and Hartos, G. R., Pollution Control and Energy Needs, Advances in Chemistry Series 127, R. M. Jimeson and R. S. Spindt, Editors, American Chemical Society, Washington, D.C., 1973, Chapter 9, "Hydro-desulfurization of Residuals", pp 98-104.

(3) Johnson, A. R., et al, ibid, Chapter 10, "H-Oil Desulfurization of Heavy Fuels", pp 105-120.

(4) Anon., "Desulfurization Refinery Capacities", Environmental Science and Technology, Vol. 7, No. 6, pp 494-496 (June 1973).

(5) Cantrell, Ailleen, Director of Editorial Surveys, The Oil and Gas Journal, Vol. 72, No. 13, pp 82-103 (April 1973).

SECTION 4

COAL REFINING AND LIQUEFACTION

Introduction

Two objectives of processes for coal refining and liquefaction are to segregate for disposal the ash, sulfur, and nitrogen impurities from the fuel or energy values, and to convert the fuel fraction to easily storable and pumpable products for use in conventional liquid fuel systems.

Refined liquid fuels have more hydrogen relative to their carbon content than does the solid coal from which they are derived. Therefore, liquefaction processes have the common feature of increasing the hydrogen/carbon ratio, either by adding hydrogen to the fuel molecule or extracting some of the carbon from the fuel in a form that can be separated from the primary liquid product.

The seven processes described in this section by individual summaries of reaction conditions, process diagrams, development history, and present status have differences that are distinguished primarily in the reactors and the reaction conditions that they are designed to accommodate. Table 4.0 outlines the features that appear to distinguish each process from the others.

Evaluation and rating of processes for use in commercial-size installations must eventually consider specific sites, available coal supplies, known process technology, production flexibility and available markets for the range of product specifications, before a choice can be made. Most of the processes described here are not developed enough for direct comparisons and recommendations. All may be suitable for large-scale operation in favorable circumstances unless current technological uncertainties require uneconomic solutions.

TABLE 4.0 COAL LIQUEFACTION PROCESSES

Process	Distinguishing Features	
	Reactor System	Other Features
4.1 H-Coal	Coal and catalyst particles suspended in circulating liquid under hydrogen	Clear liquid decanted from settling chamber
4.2 Synthoil	High velocity flow of coal/oil slurry with hydrogen through fixed catalyst bed	Unreacted solids separated by centrifuge
4.3 SRC	Dissolve coal, treat with H_2 gas, filter and precipitate coal in solution for clean, ash-free, low-sulfur fuel	Primarily fuel in utility or boiler use. Fired as pulverized solid or molten liquid.
4.4 CSF	Dissolution of coal in process solvent, with liquefaction in stirred suspension by hydrogen transfer from solvent to coal	Recovered solvent is re-hydrogenated separately before recycle
4.5 Bergius	High-pressure catalytic hydrogenation of coal slurry in oil fraction from process	Only process operated on large commercial scale (Germany, World War II) Superseded by plans for more efficient and economical processes
4.6 COED	Low temperature carbonization of fluidized bed of coal with hot gas. Adaptation of commercial carbonization processes with substitution of fluidized beds	Relatively low liquid yield compared with other processes. Appears technically proven for commercial scale
4.7 Zinc chloride molten catalyst	Zinc chloride catalyst as molten bath stirred with suspended coal or coal extract under hydrogen pressure	Large proportion of catalyst relative to coal (2:1) requires efficient separation and recycle

4.1 H-Coal Process (Hydrocarbon Research, Inc.)

Description of the Process. Dried and pulverized coal is slurried with recycled process oil and charged to a hydrogen-pressurized reactor with an ebullient bed of liquid-suspended circulating catalyst particles. Figure 4.1.1 is a diagram of the reactor in steady state operation, where the ebullient bed is maintained for uniform temperature and composition of the reaction mixture. Fluid slurry and gas are introduced from a plenum chamber through a distribution plate near the bottom. Products are removed from the top, as gas, clear liquid, and solid ash plus undissolved coal dispersed in a part of the liquid. Catalyst is sized to remain in the bed, and does not appear in the products. A small stream of catalyst suspension is removed continuously for separation, regeneration, and return to the reactor, so that catalyst activity can be maintained during continuous operation.

Figure 4.1.2 is a schematic representation of the process development unit, which has been operated to produce either synthetic crude oil or low sulfur fuel oil. Liquid yields are about 3 bbl/ton of coal on a moisture-and ash-free basis, and depend upon coal fed and product selected.

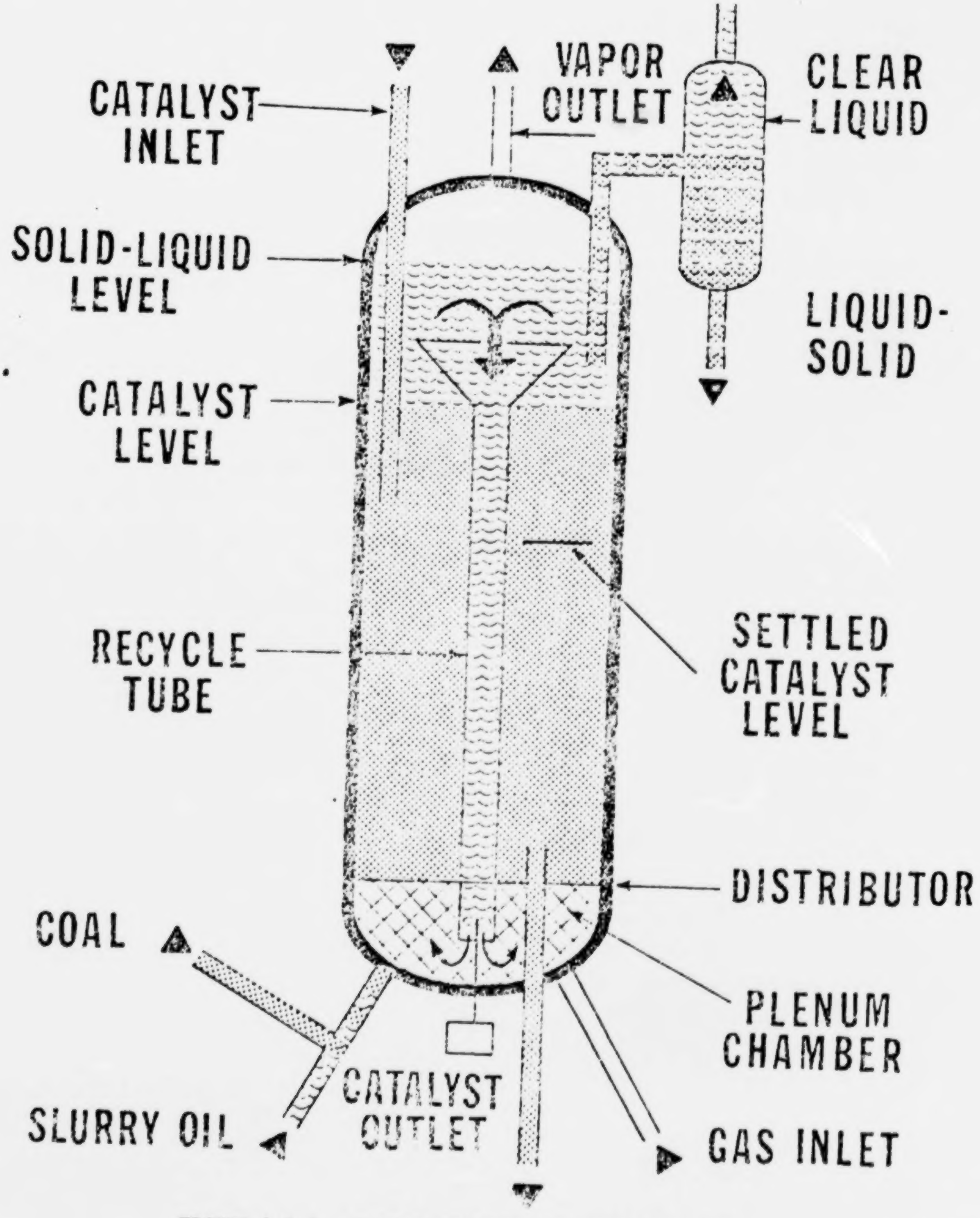

FIGURE 4.1.1. H-COAL REACTOR WITH EBULLIENT BED

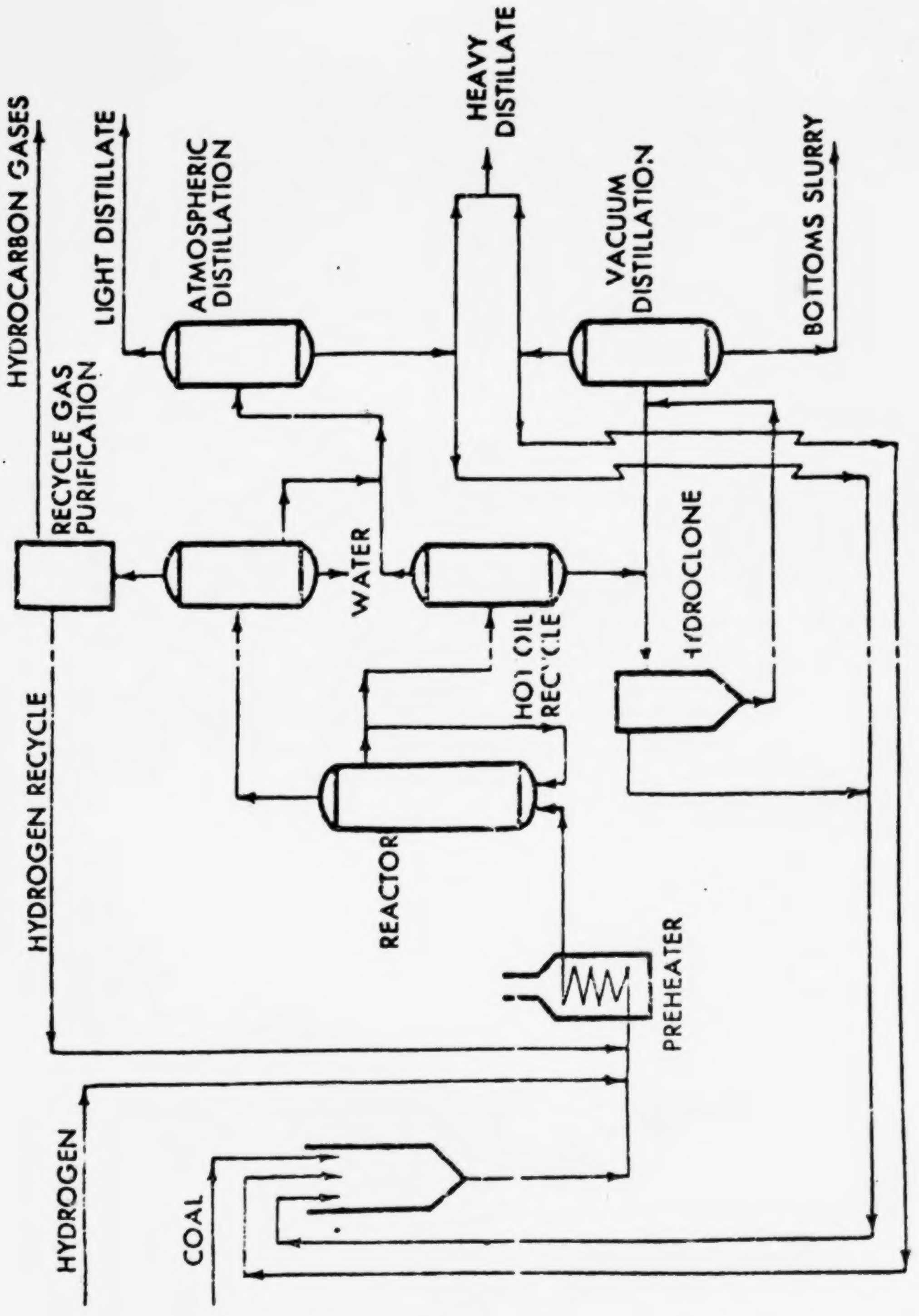

FIGURE 4.1.2 SCHEMATIC OF H-COAL PROCESS DEVELOPMENT UNIT

TABLE 4.1 H-COAL PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Bench Scale 1 lb coal/hr	Trenton, N.J.	Hydrocarbon Research Inc., 1964-1974 Initial support: OCR, 1965-1967 Industrial support 1968-1973	Data primarily from Illinois No. 6 and Wyodak coals established effect of operating variables on yields in bench scale unit (3/4" dia.)
Process Development Unit ~ 200 lb coal/hr			Process development unit confirmed bench scale results and demonstrated sustained operation with satisfactory control of ebullient bed (8-1/2" dia.).
Pilot Plant (proposed) 10-30 tons coal/hr		Pilot plant proposal to OCR 1974	Proposed (1974) pilot plant with 4.6-foot-diameter reactor to provide design data for demonstration plant with reactors 10 feet or more in diameter.

Contacts: M. C. Chervenak or C. A. Johnson, Hydrocarbon Research, Inc., Trenton, N.J. Tel 609/394-3101

References (1) Johnson, Clarence A., Chervenak, Michael C., Johanson, Edwin S., Present Status of the H-Coal Process, presented at the "Clean Fuels From Coal" Symposium, Institute of Gas Technology, Chicago, Illinois (September 10-14, 1974).

(2) Johnson, C. A., Chervenak, M. G., Johanson, E. S., and Wolk, R. H., Scale-Up Factors in the H-Coal Process, Chemical Engineering Progress (Vol. 69 No. 3) (March 1973).

(3) Johnson, C. A. and Livingston, J. Y., H-Coal: How Near to Commercialization?, presented at The University of Pittsburgh, School of Engineering, Symposium (August 6-8, 1974).

4.2 Synthoil Process (Bureau of Mines)

Description of the Process. A slurry of 35 parts coal in 65 parts recycle oil is mixed with gaseous hydrogen and injected under pressure (4000 or 2000 psi)* into a preheater (Figure 4.2) packed with ceramic pellets for efficient heat exchange. The feed to the reactor is heated to reaction temperature (450 or 430 C)* before passing into a tubular reactor containing a fixed bed of cobalt-molybdenum catalyst pellets. Throughput of slurry at the high rate of 261 lb/hr/ft ^{3} of reaction volume, together with hydrogen, induces turbulence which promotes heat transfer, helps keep catalyst surfaces clean, and prevents solids from settling out of the stream. Gas and liquid products are separated from the process stream and are further purified to remove sulfur and nitrogen. Unreacted hydrogen is recycled, and solid residues may be pyrolyzed to yield additional liquid product before being gasified to produce make-up hydrogen.

Yields from the small-scale unit (50 lb coal/hr) were about 3 bbl oil/ton coal. This unit has made continuous runs up to 500 hr before voluntary shutdown. A process development unit to react one ton of coal per hour is in the advanced planning stage. (August 27, 1974)

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* Values used in experimental tests to produce a range of product specifications.

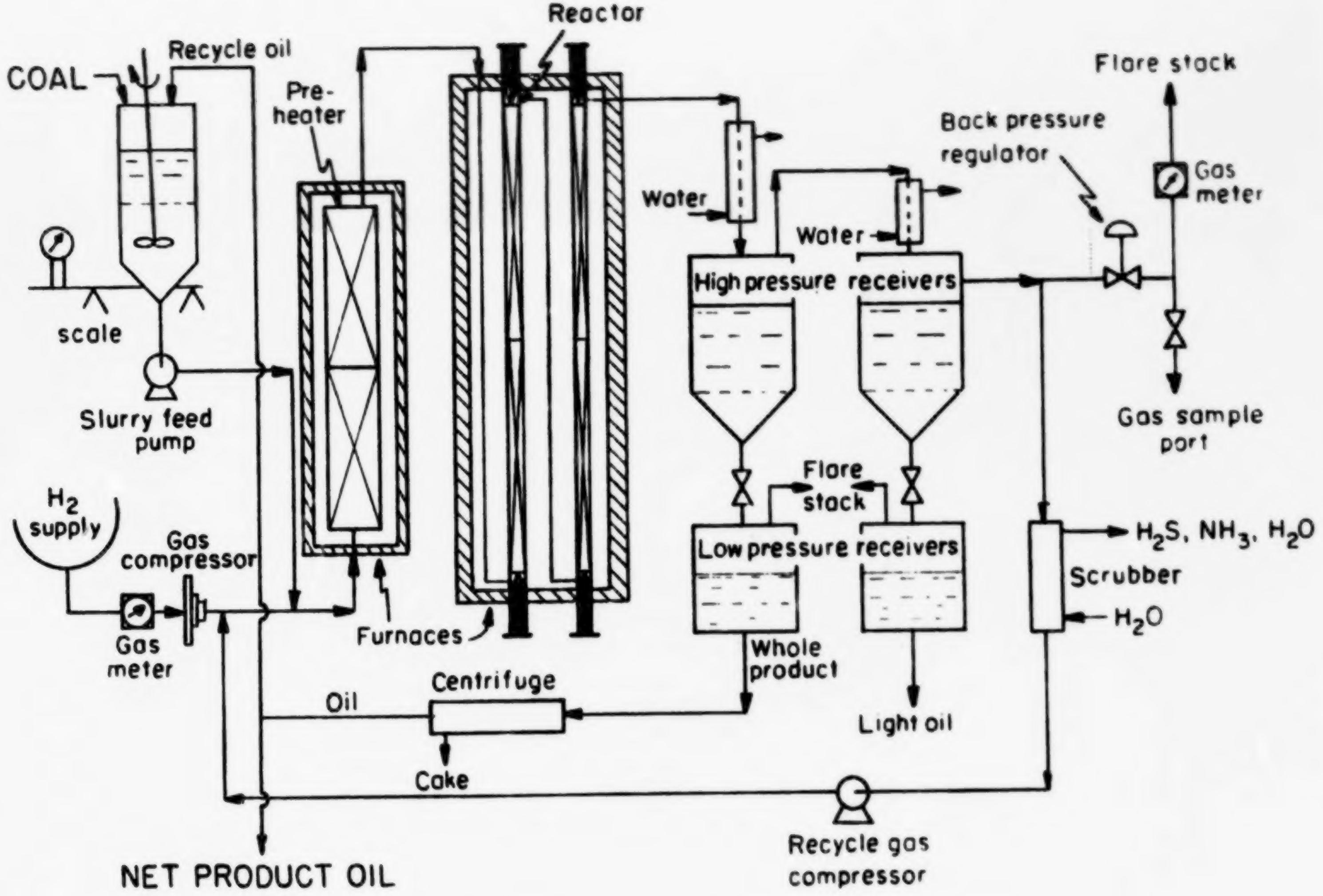

FIGURE 4.2.1 SYNTHOIL PILOT PLANT FLOW SHEET

TABLE 4.2 SYNTHOIL PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Continuous, 2 lb coal/hr	Pittsburgh, PA	Bureau of Mines, Pittsburgh Energy Research Center	This 3/16-inch tubular reactor was superseded after successful operation with up to six weeks continuous running.
15 lb coal/hr			This 1.1-inch tubular reactor is now in operation to determine yields. Runs have been completed up to 500 hours. Yield: 3 bbl oil per ton of coal. Oil specifications controlled by process conditions. Premium grade fuel oil with 0.2 percent S and <0.1 percent ash, or heavy fuel oil at lower cost. Process demonstration unit is in advanced planning stage. Favorable feasibility analysis completed. 4-inch to 6-inch dia. tubular reactor will be used.
			Comments: Accepts variety of pulverized coals. Flexibility in product specifications. No apparent attrition of catalysts. Possible by-product S and ammonia. High ash residue.

Contact: Dr. Sayeed Akhtar, Bureau of Mines, Pittsburgh, PA 15213; 412/892-2400 X306

References:
(1) Akhtar, Nestor J. Mazzocco, Weintraub, Murray, and Yavorsky, Paul M., Synthoil Process for Converting Coal to Nonpolluting Fuel Oil, presented at the 4th Synthetic Fuels from Coal Conference, Oklahoma State University, Stillwater, Oklahoma (May 6-7, 1974).
(2) Yavorsky, Paul M., Synthoil Process Converts Coal Into Clean Fuel Oil, Symposium on "Clean Fuels from Coal", Inst. Gas Technology, Chicago (Oct. 14, 1973).
(3) Yavorsky, Paul M., Akhtar, S., and Freidman, S., Converting Coal Into Non-Polluting Fuel Oil, Chemical Engineering Progress (Vol. 69 No. 3) (March 1973).

4.3 Solvent Refined Coal (SRC) Process

Description of the Process. This process is being developed to provide primarily a clean fuel suitable for firing in utility boilers and steam plants. One part of -7-mesh coal is mixed with 3 parts of a liquid fraction recycled from the product stream to form a slurry for process feed. The slurry passes through a preheater and a dissolver under hydrogen pressure of about 1000 psi. About 90 percent of the coal substance dissolves at 440 C (825 F) during a residence time of about 30 to 60 minutes (Figure 4.3.1). Undissolved solids (about 50 percent carbon) are separated by filtration. The off-gases from the reactor are washed to separate H_2S, which goes to sulfur recovery. Part of the sulfur-free gaseous hydrogen-hydrocarbon mixture is used for recycle together with make-up hydrogen, obtained by gasification of the carbon in the solid residue. Process solvent is recovered for recycle when the hot liquid filtrate is flash distilled in a vacuum column. The distillation residue is the major product. This solvent-refined coal product is low in ash and sulfur, with a softening point of about 150 C (300 F), so that it may be fired as a liquid or a solid with appropriate equipment. Gasification of the solid residue to water gas in a separate reactor is contemplated to provide makeup hydrogen to the SRC process. The water gas mixture of hydrogen, carbon monoxide, and carbon dioxide could be treated in a catalytic shift converter and washed to remove CO_2 to give hydrogen of adequate purity. An alternative has been investigated in which the water gas, fed to the process with steam, is added to the slurry as the primary process feed before entering the preheater. Results are described in progress reports to OCR from Pittsburg and Midway and are to be published.

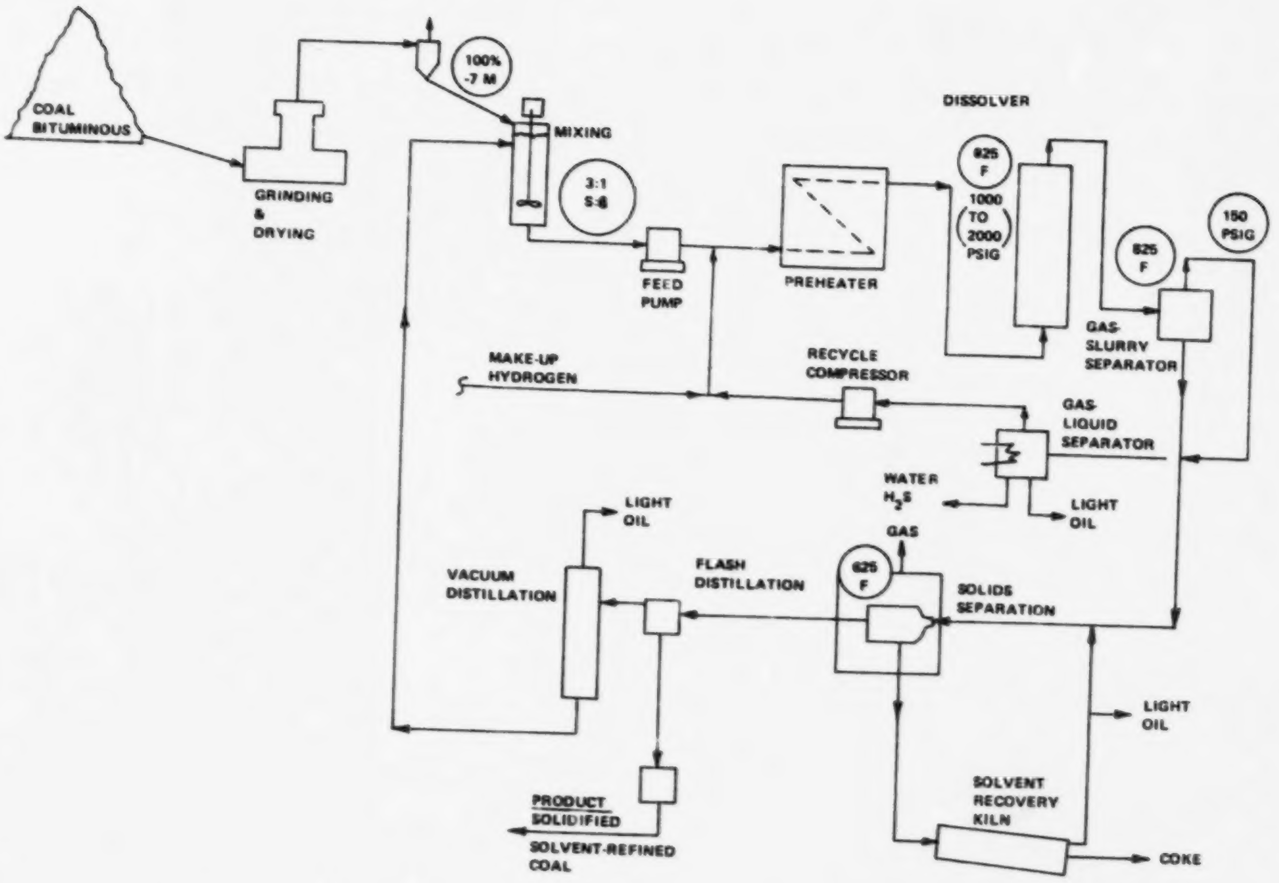

FIGURE 4.3.1 SOLVENT REFINED COAL PILOT PLANT

TABLE 4.3 SOLVENT REFINED COAL PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Bench Scale - ~ 0.5 to 1 lb coal per hour	Kansas City, MO	Pittsburgh & Midway Coal Mining Co. (OCR)	Bench scale studies from 1962 to 1966 and 1968 to present to study product properties and solubility rates as functions of operating parameters.
Pilot Plant - 2 tons coal per hour	Fort Lewis, WA	"	Construction completed and components undergoing performance tests in September, 1974. Continuous integrated operation not yet achieved.
Pilot Plant - 500 lb coal per hr	Wilsonville, AL	Southern Services, Inc. (EEI, Southern Co.)	Intermittent operations at less than rated capacity since January, 1974. One continuous test for 45 days. Estimated availability of quantities of light oil product for evaluation in " a month or two". (September, 1974). Some reported difficulties with filtration step being below rated capacity.

Contacts: Everett L. Huffman, Southern Services, Inc. Box 2625 Birmingham AL 35203 Telephone 205/870-6324

Bruce K. Schmid, Pittsburgh and Midway Coal Mining Company, Ft. Lewis, WA Telephone 206/964-8155

Reference: B. K. Schmid, "The Solvent Refined Coal Process", Symposium on Coal Gasification and Liquefaction, Univ. of Pittsburgh, Pittsburgh, PA., (August 6-8, 1974)

4.4 CONSOL Synthetic Fuel (CSF) Process

Description of the Process. Crushed and dried coal (1 part) is mixed (Figure 4.4.1) with a process middle-fraction recycled solvent (2.5 parts) and dissolved at process temperature (400 C). Solvent is prepared for recycle by catalytic hydrogenation and serves as a hydrogen donor during liquefaction of 70 to 85 percent of the raw coal in a stirred reactor under solvent vapor pressure for 30 minutes. Hydrogen transferred from donor to coal is about 0.7 weight percent of the coal. Unreacted coal and ash are separated by filters, hydroclones, centrifuges, or a combination thereof. The oil product would be suitable as utility fuel or as low-sulfur feed to an oil refinery.

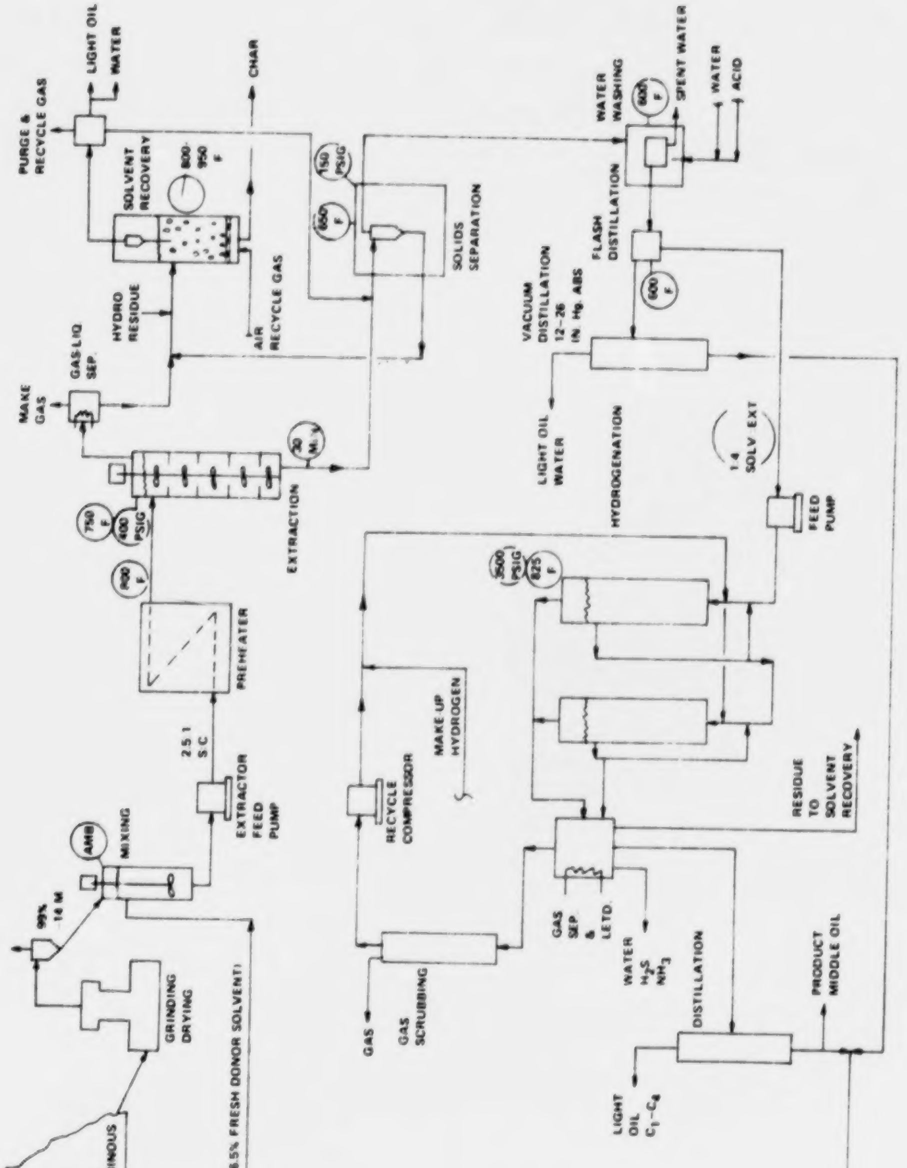

FIGURE 4.4.1 CONSOL SYNTHETIC FUEL PROCESS. 1 TON PER HOUR PILOT PLANT FOR CRUDE OIL PRODUCTION

TABLE 4.4 CONSOL SYNTHETIC FUEL (CSF) PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Bench scale - 20 lb coal/hr	Library, PA	Consolidation Coal Company (OCR "Project Gasoline") 1964-1970	Bench scale provided design data for pilot plant
Pilot Plant, 1 ton coal/hr	Cresap, WV		Pilot plant construction started 1967. Recurring mechanical and operational difficulties with about 500 hours total operation until shutdown in April, 1970.
			Three year contract to revamp, modify, (18 months, standby to July, 1974 thru Dec. 1974) and operate (18 months) plant for for liquefaction data to design demonstration plant.
		Fluor Corporation, contractor (OCR, Am. Electric Power Co., Allegheny Power Co.)	Plans for modification and operation include components testing, integration, and possibly three modes of operation for production of liquid fuel: (1) hydrogenation of extract as hydrogen donor, (2) fixed catalyst bed to hydrogenate coal + recycle oil slurry, (3) ebullated catalyst bed with coal, recirculated oil, and hydrogen.

Contact: Harold L. Finch, Fluor Corporation, Cresap, WV, (near Moundsville, 26041) Telephone 304-845-2211 August 27, 1974

References: (1) J. A. Phinney, "Clean Fuels via the CSF Process", Symposium on Clean Fuels from Coal, Institute of Gas Technology, Chicago, IL October 10-14, 1973.

(2) Press Release, "OCR's West Virginia Pilot Plant to Test Coal-to-Liquid Process Components", Office of Coal Research, Washington, D.C. June 7, 1974.

4.5 Bergius Coal Liquefaction Process

Description of the Process. Germany used the bergius process to produce gasoline and lubricating oils on a commercial scale during World War II by catalytic hydrogenation of brown coal. Figure 4.5.1 shows a flow diagram of the process. After the war, the United States Bureau of Mines operated a pilot plant at Louisiana, Mo,* to study the direct hydrogenation-liquefaction of American bituminous coals by the Bergius process as well as modifications for improved efficiency. Improvements were incorporated into the process, and operation was generally successful. Work was discontinued in the early '50's because the process was uneconomic as long as petroleum was available. There are no active studies of the Bergius process now (1974).

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* H. R. Batchelder, "Synthetic Fuels, Economics, and Future Trends", Chapter I, Advances in Petroleum Chemistry and Refining, Vol. 5 pp 1-77 (1962).

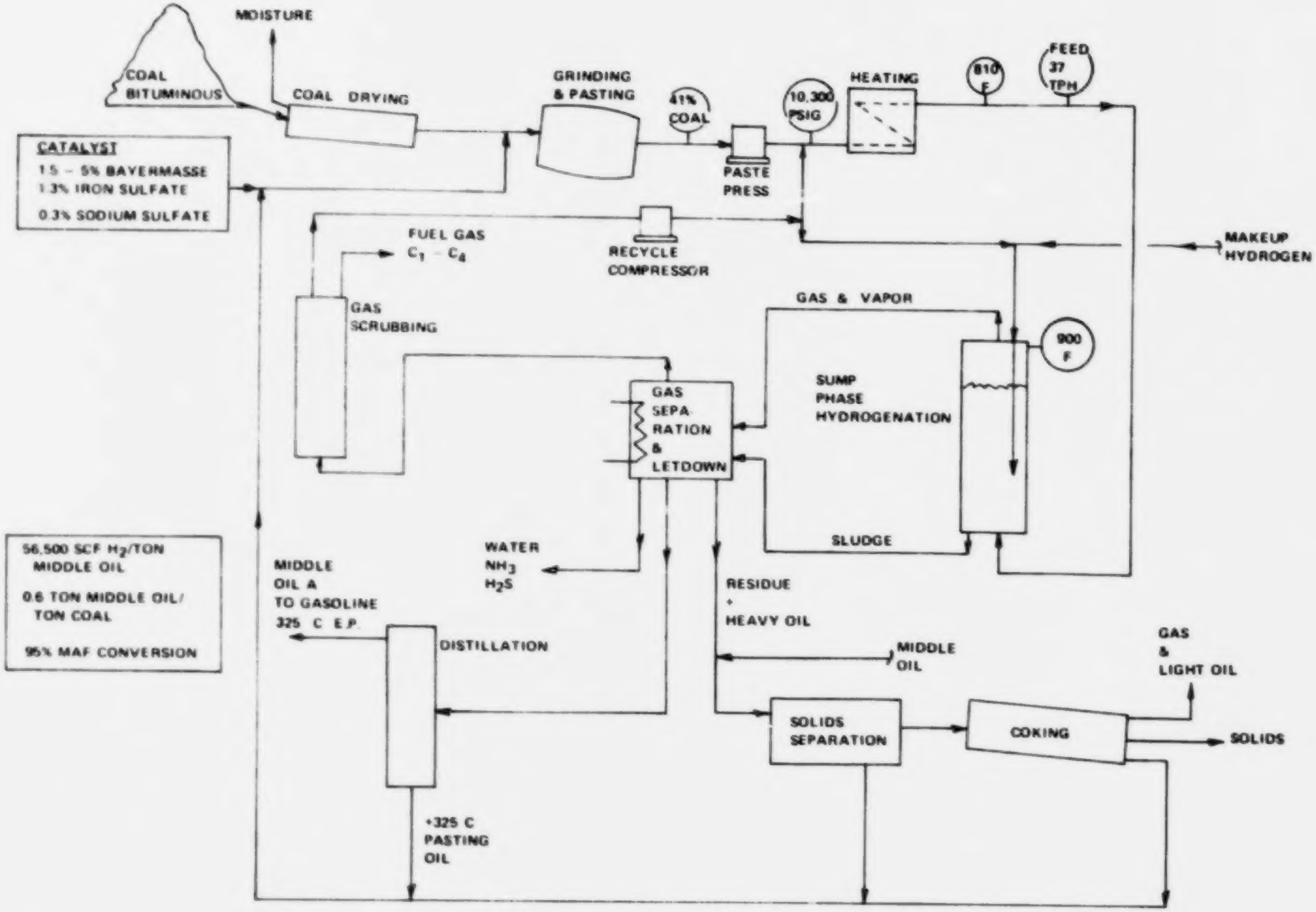


FIGURE 4.5.1 BERGIUS CATALYTIC HYDROGENATION GERMAN COMMERCIAL PRACTICE

4.6 COED Process (FMC Corporation)

Description of the Process. The COED process carbonizes coal in a series of four fluidized bed reactors by direct contact with heated, fluidizing gas. Gases pass successively from the higher to lower temperature beds as the carbonized char effluent flows countercurrent to the gas from low to higher temperatures. Bed temperatures are controlled to dry, devolatilize, and pyrolyze caking coals without softening and agglomeration (Figure 4.6.1). Flue gas flows through the first stage at 300 C (550 F), to a drier for the incoming coal, and through a clean-up section to the stack. Steam and oxygen injected into the fourth stage are controlled to maintain a temperature of 850 C (1550 F) by burning a part of the char. The gases and volatiles from the fourth stage flow successively through the third stage at 565 C (1050 F), the second stage at 450 C (850 F), and thence to the oil recovery unit where gases are separated for purification and liquids proceed to a hydrogenation unit to produce a synthetic crude oil suitable for refining. The major products and approximate yields per ton of coal are: liquid syncrude, 1 barrel; 600 Btu fuel gas, 8000 SCF; char, 1200 lb.

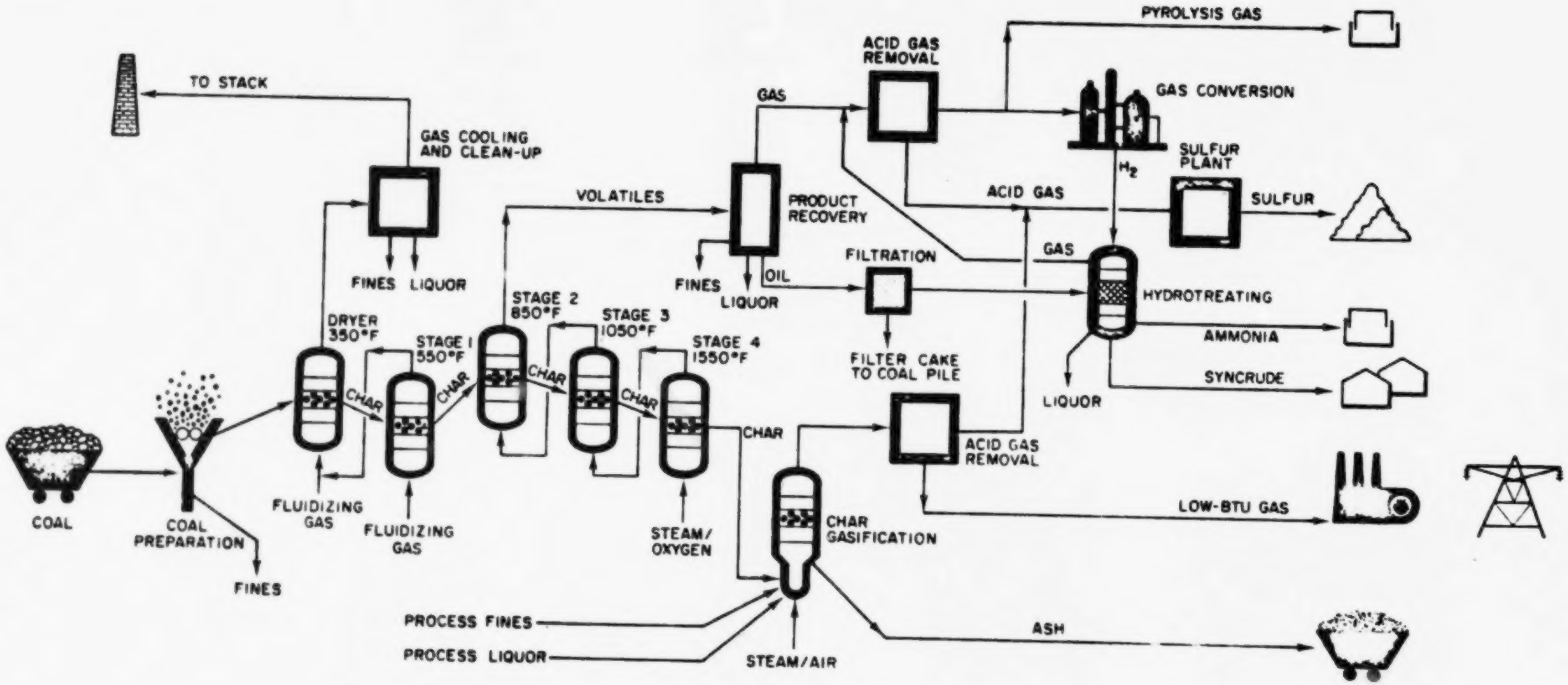


FIGURE 4.6.1 COED PROCESS WITH ADDED CHAR GASIFICATION FMC-OCR

TABLE 4.6 COED PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Bench Scale and Process development Unit (100 lb coal per hr)	Princeton, NJ	FMC Corp. (OCR)	Bench scale research and PDU operated 1962 to 1970 to provide data for pilot plant.
Pilot Plant 1.5 tons coal/hr	"	"	1970 to present. Six typical coals and lignite processed in integrated operations up to 30 days. Two-week runs in hydrotreating section. Fully operational. Total coal processed has been 18,000 tons. Sufficient data for design of demonstration plant.
Demonstration plant 25,000 tons coal per day	"	"	Conceptual design and economic study completed 1974

Contact: N. J. Brunsvold, Manager, Commercial Development, COED Process, FMC Corporation, Box 8, Princeton, NJ 08540
Telephone 609/452-2300 July 29, 1974

References (1) Hamshar, J. A., Terzian, H. D., Scotti, L. J., Clean Fuels From Coal By The COED Process, for presentation at Environmental Protection Agency Symposium, St. Louis, Missouri (May 1974).

(2) Scotti, L. J., Jones, J. F., Ford, L., McMunn, B. D., Multi-Stage Fluidized-Bed Pyrolysis of Coal at the Project COED Pilot Plant, for presentation at American Institute of Chemical Engineers 77th National meeting, Pittsburgh, Pennsylvania (June 2 to 5, 1974).

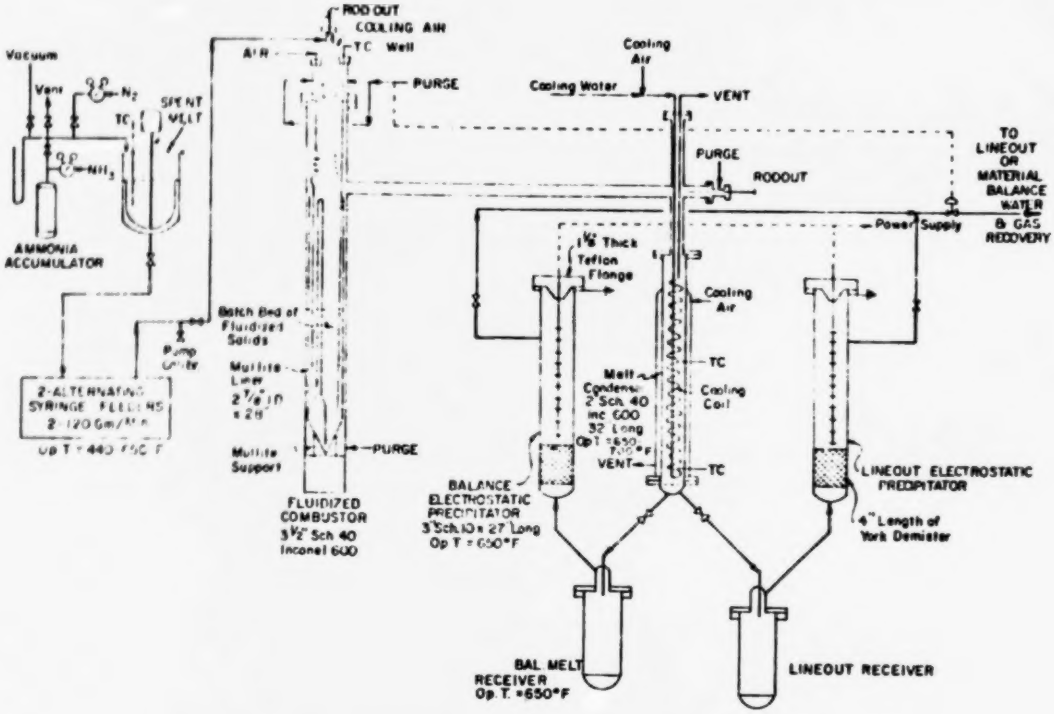
(3) Scotti, L. J., "The COED Process - Technology and Economic Feasibility", University of Pittsburgh Symposium (August 6-8, 1974).

4.7 CONSOL $ZnCl_2$ Process

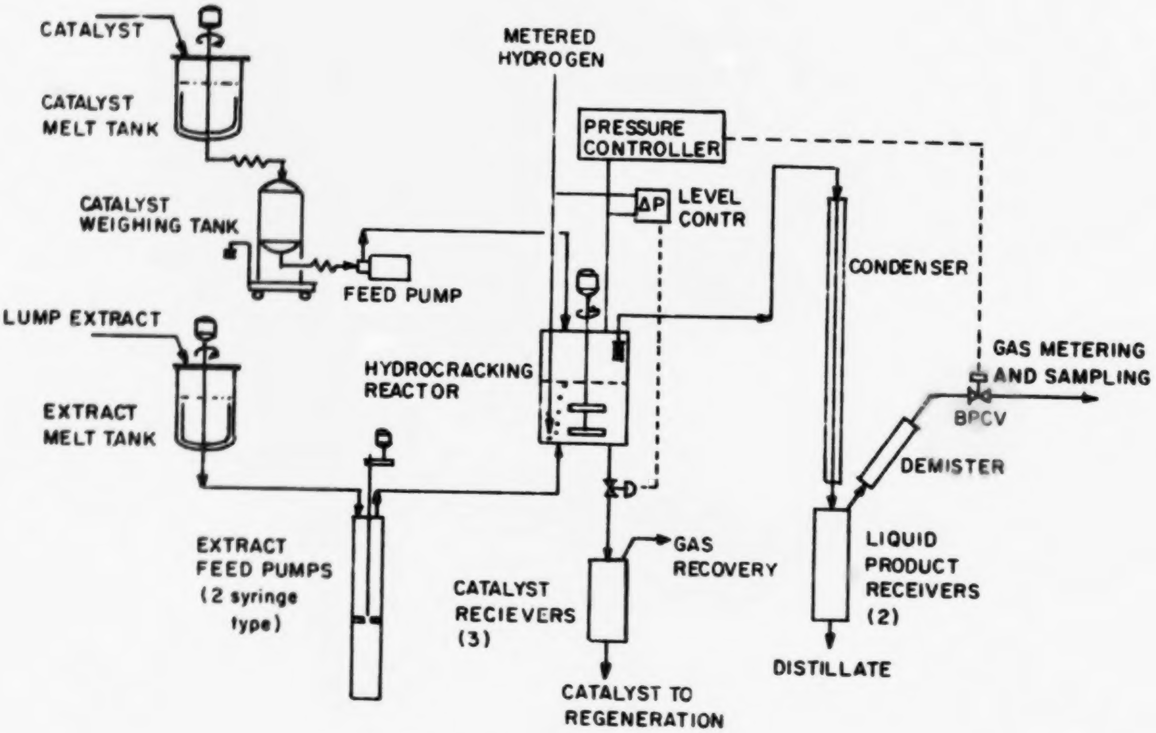
Description of the Process. Molten zinc chloride is used as a hydrocracking catalyst mixed with a tetralin extract of coal or with untreated coal in about 2:1 proportions of catalyst and coal substance. The reaction mixture is maintained in a stirred reactor under hydrogen pressure of 100 to 200 atmospheres at temperatures of 360 to 425 C. Specific conditions are selected to control the proportions of gasoline and low-sulfur fuel oil, which are the major products. The process has been operated on a bench scale in continuous units for hydrocracking (Figure 4.7A) and for regeneration of the spent catalyst (Figure 4.7B) by combustion of retained impurities in a fluidized bed of inert particles. Most of the work was carried out between 1964 and 1967 under sponsorship of the Office of Coal Research. Recent studies of methods for regeneration of the spent catalyst have been made by Consolidation Coal Company. It is reported that a resumption of work on a larger scale is under consideration for renewed support by the Office of Coal Research.

A unique advantage of this process is the demonstrated capability to convert a large part of the coal substance directly into naphtha boiling in the gasoline range and having a high octane number. Reaction conditions are relatively mild and residence time in the reactor is reasonably low.

The major disadvantage is the use of massive quantities of zinc chloride catalyst which must be purified and recycled economically to arrive at a commercially viable process.



B. Catalyst Regeneration System



A. Hydrocracking Process

FIGURE 4.7. CONSOL $ZnCl_2$ PROCESS

TABLE 4.7 CONSOL ZINC CHLORIDE PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Bench Scale Hydrocracking, Batch Autoclave 300 ml.	Library, PA	Consolidation Coal Company (OCR)	Reaction rates and conversions determined on bench scale equipment 1964-1967.
Continuous stirred reactor, 500 g inventory			In-house studies of catalyst regeneration, 1973.
Catalyst regeneration, continuous feed at 2-120 g/min.			Process demonstration unit under consideration for OCR support, 1974.

Contact: **Dr. Everett Gorin, Research Division, Consolidation Coal Company, Library, PA 412/288-8700**

References:

(1) Zielke, C. W., Struck, R. T., Evans, J. M., Costanza, C. P., Gorin, E., Molten Zinc Halide Catalysts For Hydrocracking Coal Extract and Coal, I&EC Process Design and Development, Vol. 5 pp 158-164 (1966).

(2) Struck, R. T., Clark, W. E., Dudt, P. J., Rosenhoover, W. A., Zielke, C. W., Gorin, E., Kinetics of Hydrocracking of Coal Extract with Molten Zinc Chloride Catalysts in Batch and Continuous Systems, I&EC Process Design and Development, Vol. 8 pp 546-551 (1969).

(3) Zielke, C. W., Struck, R. T., and Gorin, E., Fluidized Combustion Process For Regeneration of Spent Zinc Chloride Catalysts, I&EC Process Design and Development, Vol. 8 pp 552-558 (1969)

(4) Zielke, C. W., Rosenhoover, W. A., Gorin, "Direct Zinc Chloride Hydrocracking of Subbituminous Coal--Regeneration of Spent Melt", Preprints Volume 19, No. 2, pp 306-311, Division of Fuel Chemistry, Am. Chem. Soc., 167th National Meeting, Los Angeles, CA (April, 1974).

SECTION 5

LOW-AND INTERMEDIATE-BTU GAS FROM COAL AND OIL

Introduction

Gaseous products have been generated from coal for more than 150 years, and,although gas from coal was once used extensively for commerical and residential lighting and cooking,it has never been a significant factor in the U. S. energy economy. That situation is likely to change within the next few decades. Increasing costs and decreasing supplies of clean fuels coupled with the existence of extensive domestic reserves of coal has led to a dramatic resurgence in coal gasification research and development.

Gasification of coal transforms an abundant but inconvenient dirty solid fuel into a convenient and clean gaseous fuel. In general, the process involves reaction of coal with air, oxygen, or steam,or mixtures of these gases to yield a combustible product containing carbon monoxide, carbon dioxide, hydrogen, methane, and nitrogen.

Low-Btu gas (about 125-175 Btu/SCF) is obtained when coal is gasified with air-steam mixtures with the result that nitrogen is the major component of the product gas. Because of its low heating value, it cannot be economically stored or pipelined over long distances. Nonetheless, there are many potential uses near the source of production including power generation, steam generation,and industrial heating. Such uses are likely to be especially significant in areas such as the East and Mid-west where most of the coal is relatively high in sulfur.

Although intermediate-Btu gas (about 300-500 Btu/SCF) is usually produced by gasification with steam and oxygen, there are also processes in which pure oxygen is not required (e.g., the CO_2-Acceptor Process and the Union Carbide-Battelle Ash Agglomeration Process). In the latter processes the heat necessary for gasification is provided by some method other than reaction of oxygen and carbon.

Processes in this chapter are classified on the basis of the type of fuel-bed. They range in degree of development from those that have not yet reached the pilot stage to those such as the Lurgi, the Winkler and the Koppers-Totzek that have been operated on a commercial scale for a number of years. Some of the advanced design gasifiers described in this chapter are primarily intended for integration into processes for producing high-Btu gas (Section 6). They are included here because they can be operated to produce intermediate and/or low-Btu gas. It should be noted that many of these advanced design gasifiers are designed to operate at very high pressures (in excess of 1000 psi) in order to increase direct yields of methane. Such high pressures are unnecessary in the production of a low Btu-fuel gas.

Underground (in situ) gasification and the gasification of high-sulfur refinery residues are also considered.

References to each specific process are given at the foot of the respective "State-of-the-Art" table. General references to coal gasification are given on Page 6-37.

Many of the gasification processes developed prior to 1960 have been omitted because they are not presently receiving serious consideration for the production of fuel gases. Several excellent reviews of these earlier processes are available in the literature.(*)

At this stage of development, environmental factors are best treated in rather general terms. Moreover, because of the considerable overlap in the environmental aspects of various gasification systems, it is convenient to discuss all of the processes in this chapter and those in Section 6 (Pipeline-Quality Gas) together. This discussion is presented in Appendix A.

(*) Newman, L. L., Industrial and Engineering Chemistry, 40(4), 559-81 (1948); Shires, G. L., Chem. Eng. and Mining Rev., pp 43-50 (Aug. 15, 1958), pp 41-47 (September 15, 1958).

Status of the Technology

Interest in the production of low- and intermediate-Btu fuel gases has developed comparatively recently. The most attractive application is in an integrated gasification-power plant complex. The clean gas may be used as a fuel for either a conventional steam generating plant or in one of the advanced power cycles, which are potentially much more efficient (41). The combined (gas turbine/steam turbine) cycle is the most highly developed of the advanced cycles and is thus the one most frequently considered for eventual integration with a coal or oil gasification plant. Figure 5.0 illustrates a possible configuration for such an integrated system.

A demonstration plant is now in operation in West Germany which generates about 170 MW of power from low-Btu gas produced from coal in Lurgi gasifiers (Table 5.1.1). Presently, the overall thermal efficiency is only 36 percent; however, this could be increased substantially by increasing pressure levels and turbine inlet temperatures.

In the U.S., a number of more advanced gasification processes are being developed for integration with combined-cycle power plants. In particular, Foster-Wheeler Corporation is expected to begin construction soon on a 50 TPH pilot plant which in phase I operation will provide low-Btu gas to modified existing boilers. In phase II, projected to begin in early 1978, the low-Btu gas will fuel a combined-cycle plant to produce about 130 MW of power. Westinghouse has a 1200 lb/hr process development unit of its fluidized-bed gasifier in the pre-commission stage. If significant success is achieved in tests with this unit, they may go directly to a full-size pilot integrated with a combined-cycle power plant rated at about 120 MW. These projects are funded by OCR.

Texaco Oil and United Aircraft have carried out pilot-scale studies of the combustion of low-Btu gas in an existing gas turbine system. The gas produced from oil (13 API, 650 F initial B.P., 2 percent S) by Texaco's Partial Oxidation Process, was burned in a Pratt & Whitney FT 4 combustor test stand (43). Results demonstrate that low-Btu gas can be

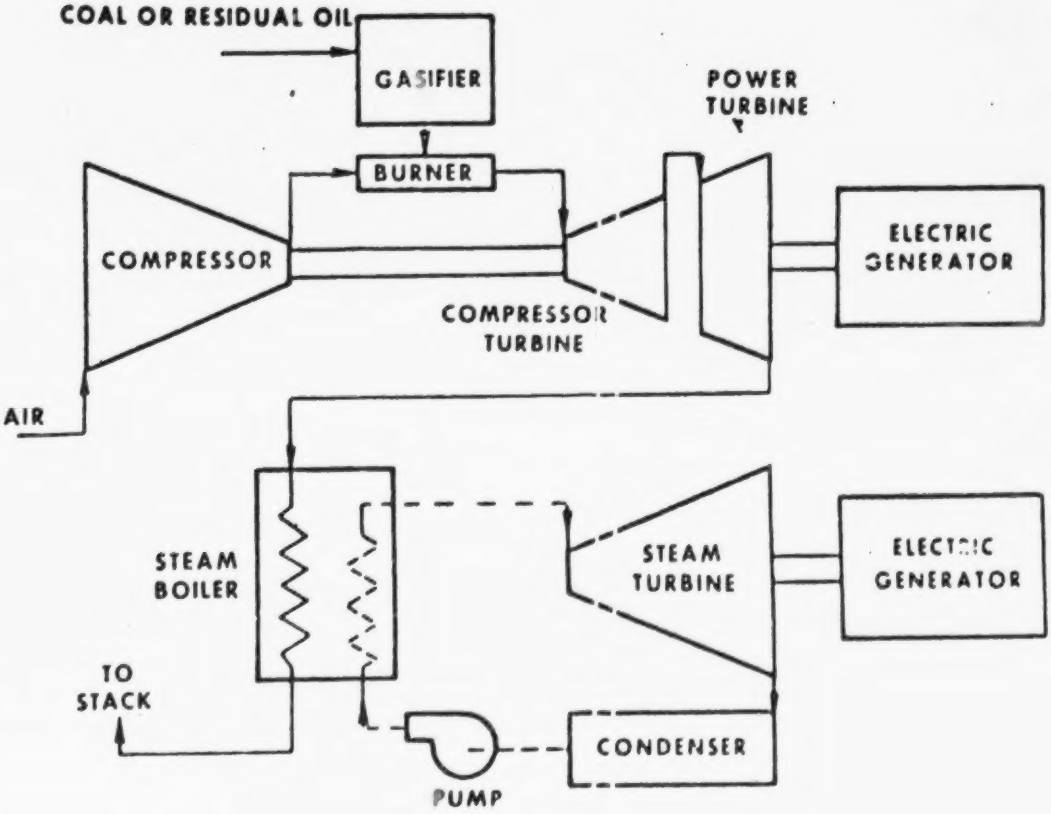


FIGURE 5.0. GASIFIER - COMBINED CYCLE COMPLEX

efficiently burned, and necessary gas turbine modifications are described. The Texaco gasifier can also accept coal and high-sulfur residual oils.

Other gasification processes being developed for integration with combined cycle power plants include the following: Gegas (General Electric), U-Gas (IGT), COGAS, Combustion Engineering, Kellog Molten Salt, and Atomics International Molten Salt.

Probably the most demanding requirement for gasification processes integrated with combined-cycle systems is gas purity. Current specifications require that particulates be reduced to 0.08 lb/MMCF or less. No particulate size is specified, but to minimize blade erosion few particles should be larger than 20 or 25μ (41).

Total sulfur in the fuel is limited to 162 lb/MMCF of which hydrogen sulfide can be no more than 0.18 percent (vol.).

Ideally, the gas should be purified while it is still at high temperatures, but most present methods require the gas to be cooled to a relatively low-temperature (250 F or lower). Several hot-gas purification processes are being investigated.(*) Development of a successful process would lead to a significant increase in thermal efficiency.

In the more immediate future, low- and intermediate-Btu gas will probably find use as a fuel in modified existing boilers. Although it will not be possible in all cases to retrofit existing boiler to burn low-Btu gas (27), this process could free considerable quantities of natural gas for other uses.(**) Recent studies (26) suggest that low-Btu gas from coal may compete favorably with stack-gas scrubbing in coal burning plants.

Commonwealth Edison of Chicago is installing three Lurgi gasifiers to produce 150-200 Btu/SCF fuel gas from coal. The gas will be burned in modified existing boilers at its Powerton Station in Pekin, Illinois (Table 5.1.1); and OCR and TVA plan to install two or more stirred-bed (USBM) gasifiers at a TVA power plant for a similar purpose (Table 5.1.3).

Gasifiers are also likely sources of low- and intermediate-Btu gas for industrial use. Several Wellman-Galusha gasifiers are currently

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(*) Among these are Exxon's process using dolomite in a fluidized bed, Battelle's molten salt process, the Bureau of Mines iron oxide process and IGT's Meissner process.

(**) More than 65 percent of the natural gas sold in this country is consumed by the electric utility industry and industry generally.

being used for this purpose (Table 5.1.2) and more wide-spread use of gasifiers can be expected as supplies of natural gas become tighter.

The current status of each of the processes considered in this Section is described in individual state-of-the-art tables that follow; a summary of this information is given in Table 5.0.
An excellent and relatively recent evaluation of the technology for producing low- and intermediate-Btu gas from coal appears in Reference 8; the status of in situ gasification is appraised in Reference 45.

It is also useful to consider some of the more general problems associated with unit operations common to most of the processes for generating low- and intermediate- Btu fuel gases. Among these are the following:

Coal Mining. The development of advanced surface and underground mining technology that will increase productivity and coal recovery and at the same time meet health, safety and environmental standards will have an important bearing on coal gasification.

Coal Preparation. At present it is not possible to crush or grind coal to a specific size without production of surplus fines. In most of the fluid-bed processes the coal must be sized to prevent high fuel losses due to carbon-carry over, and most fixed-bed processes cannot accept fines.

Coal Feeding. Because operating pressures are generally rather moderate, typically a few hundred psi, feeding coal to the gasifier is not nearly as great a problem as when SNG is being produced (pressures often 1000 psi or greater). Nonetheless, considerable development effort in this area is justified.

Refractory Problems. Under conditions prevailing in many gasifiers, SiO_2 reacts with hydrogen to produce SiO and water vapor. This reaction is reversed downstream, where temperatures are lower and pipes can become clogged with SiO_2. Presently, the approach is to use expensive alumina refractries (39).

TABLE 5.0. STATUS OF LOW AND INTERMEDIATE-BTU GASIFICATION: A SUMMARY

Process/Developer	Current Status	Comments
Moving-Bed/Dry Ash:		
Lurgi	Commercial	Because the maximum size of the gasifier is limited, several gasifier units must be operated in parallel. Older models accepted only noncaking coals. Modified version has been tested successfully with caking coals. See Table 5.1.1.
Wellman-Galusha	Commercial	Standard gasifier accepts only anthracite or coke. Agitator model also accepts bituminous coal. See Table 5.1.2.
U.S. Bureau of Mines*	1200 lb/hr PDU, Morgantown, W.Va.	OCR and TVA plan to install two or more commercial-size gasifiers designed on the basis of this unit. Tests indicate that it can accept strongly caking coals.
Gegas/General Electric	50 lb/hr unit in operation since 1971; 1200 lb/hr PDU is in the design stage.	Hope to develop a unique coal extrusion process for feeding coal to the gasifier. Also developing membrane systems for gas clean-up. Process will be used to produce low-Btu gas for power plants.
Kellog	4 TPH pilot should be in operation in mid-1975; Houston, Texas	
Moving Bed/Slagging:		
Thyssen-Galocsy	Defunct	40 TPD pilot plant was operated in Germany in 1943-44. Work interrupted by WW II and not resumed.
Fluid-Bed/Dry Ash:		
Winkler	Commercial	See Table 5.3.1
Synthane*/US Bureau of Mines	3 TPH pilot under construction at Bruceton, Pa. Expect completion in Jan. 1975.	Studies with a 40 lb/hr gasifier indicate process can accept any U.S. coal.
CO_2 Acceptor*/Consolidation Coal	1.5 ton/hr pilot in operation since 1972 at Rapid City, S.D.	Can only accept lignite and subbituminous coal. Problems with refractory failure and plugging of acceptor lines during start up seem to be solved.
Exxon Oil Company	20 ton/hr pilot under construction at Baytown, Texas	Process will be used to produce intermediate Btu gas for upgrading to SNG.
Hydrocarbons Research Inc.	10 ton/day process development unit available in Trenton, N.J.	First operated in 1958 with anthracite. Modified in 1972 to accept bituminous coal.

TABLE 5.0 (Continued)

Process/Developer	Current Status	Comments
COGAS/Cogas Development Company	4 ton/hr pilot in operation since March 1974, in Leatherhead, England. Also, a 400 lb/hr pilot has been in operation in Plainsboro, N.J. since May 1974.	Process will be used to produce intermediate-Btu gas for upgrading to SNG.
Bituminous Coal Research*	Bench-scale	$2,575,000, 50-month contract with OCR to cover bench-scale and PEDU work that will provide the basis for design of a pilot plant. Process will be used to produce low-Btu gas for power plants.
Fluidized Bed/Agglomerating Ash:		
U-Gas/IGT	10 ton/hr pilot is in design stage.	Seeking funding for pilot plant. Present studies are being made with a 4 ft. diameter gasifier. Primary purpose is to provide low-Btu gas for power plants.
Westinghouse*	1200 lb/hr PDU mechanically complete; expect first hot tests in January 1975; Waltz Mill. Pa.	If significant success is achieved with this unit, a decision could be made to go directly to a full-size pilot integrated with a combined-cycle power plant (120 MW).
Ash Agglomeration*/ Battelle-Union Carbide	1200 lb/hr PDU under construction at W. Jefferson, O. Estimate completion in first quarter 1975.	
Entrained Flow:		
Bigas*/Bituminous Coal Research	5 ton/hr pilot under construction at Homer City, Pa. Estimate completion in early 1975.	
Combustion Engineering*	5 ton/hr pilot.	Presently negotiating contract for construction. Vefy little information available at this time. Process will be used to produce low-Btu fuel gas for power plants.
Foster-Wheeler*	50 ton/hr pilot. Construction is expected to begin in fourth quarter of 1974.	In phase I the pilot plant will provide low-Btu gas for modified existing boilers. In phase II (early 1978) the gas will fuel a combined-cycle power plant.

TABLE 5.0 (Continued)

Process/Developer	Current Status	Comments
Garret Flash Pyrolysis/ Garret Research & Development Co.	10 ton/hr pilot plant has been proposed.	Seeking support. A bench-scale unit (50 lb/hr) has been in operation since Jan. 1973.
Entrained Flow/Slagging:		
Koppers-Totzek	Commercial	See Table 5.6.3.
Texeco	Not presently operational	Texaco has previous pilot plant experience and a semi-commercial unit was in operation for a number of years.
Babcock & Wilcox - DuPont	No longer in use.	A 17 ton/hr commercial unit was operated for about one year in the early fifties by DuPont at Belle, West Virginia. Dismantled.
Molten Bath:		
Molten Iron/Applied Technology Corp.	Bench-scale.	Studies have involved a 25 inch I.D. induction furnace to simulate the gasifier (4,000 lb capacity).
MoltenSalt/M. W. Kellog Co.	Bench-scale.	A process development unit is planned and preliminary flow sheets and cost estimates have been made.
Molten Salt/Atomics International	Bench-scale.	Process will be used to produce low-Btu fuel gas for power plants.
Underground gasification*	Pilot scale tests at Hanna, Wyo.	See Table 5.8.
Gasification of Residual Oils:		
Flexicoking/Exxon Corp.	Commercial	See Table 5.9.1.
Texaco Partial Oxidation/Texaco Oil Co.	Commercial	See Table 5.9.2.
SRP/Shell Oil Company	Commercial	See Table 5.9.3.

TABLE 5.0 (Cont'nued)

Process/Developer	Current Status	Comments
H-Gas/Hydrocarbons Research Inc.	Laboratory-scale.	Studies with a lab scale unit capable of processing up to 15 B/D demonstrate that resid and heavy sour crudes can be gasified.
IGT	Laboratory-scale.	Studies of the kinetics of oil char gasification. Economic analysis of the process.

*Process is currently funded in whole or inpart by OCR or US Bureau of Mines.

Hot Char Transport and Injection. Many of the new processes require the recycling of char. Development of a system capable of transporting and injecting char at high temperatures and pressures without producing serious erosion is a problem that requires much more work.

Quench Chamber Corrosion. In many cases, the gasifier effluent is quenched to scrub out heavy hydrocarbons and particulates. Because of the high temperatures and the corrosive nature of the gas (relatively high concentrations of hydrogen sulfide), materials for this stage of the system are subject to severe corrosion. Considerable research is still needed on materials for this stage (39).

Gas Clean-up. Substantial improvements in overall efficiency could be realized if processes can be developed that will clean the gas at high temperatures. A number possible methods are presently being investigated.(2)

5.1 Moving Bed/Dry Ash

In moving-bed gasifiers the fuel bed is supported by a grate and maintained at constant depth. Fuel moves slowly downward through various zones in the bed and solid residue (dry ash) is discharged from the bottom. Because of the counter-current flow of gas and fuel, heat economy is excellent, and the relatively long residence time of fuel in the reactor leads to high carbon conversion. The unit processes are relatively simple and the technology is well-developed; however, this type of gasifier generally has been operated only with non- or weakly-caking coals and sized fuel must be used for maximum output. Methods of easing or eliminating these restrictions on the coal feed are being investigated and results will be considered in this section.

5.1.1 Lurgi (The American Lurgi Corp.)

Description of the Process. The Lurgi gasifier (Figure 5.1.1), which is water-jacketed, operates at pressures of 300-500 psi. Lump coal (1 inch to 28 mesh) is fed through a lock hopper system and distributed by a rotating arm. Oxygen (or air) and steam are fed through the grate to gasify the coal and ash is removed by the rotating grate through an ash lock on the bottom of the unit. Excess steam is added to keep the ash from slagging. The Lurgi gasifier combines hydrogasification and production of synthesis gas in one reactor: synthesis gas generated in the lower region of the gasifier by reaction of steam and oxygen with coal passes up through the coal bed to devolatilize, dry, and hydrogasify the coal. Bottom temperatures are about 1800 F and the crude gas leaving the gasifier is at about 1100 F.

The raw gas undergoes direct spray washing with water to partially cool and clean the gas. Following further cooling in a waste-heat boiler, remaining tars and oils are removed by a water-cooled condenser. Tars and oils are separated and recycled to the gasifier.

Finally, hydrogen suflide and carbon dioxide are removed by a purification process, such as Rectisol, to yield a product with a heating value of about 450 Btu/scf if the gasifier is oxygen blown.

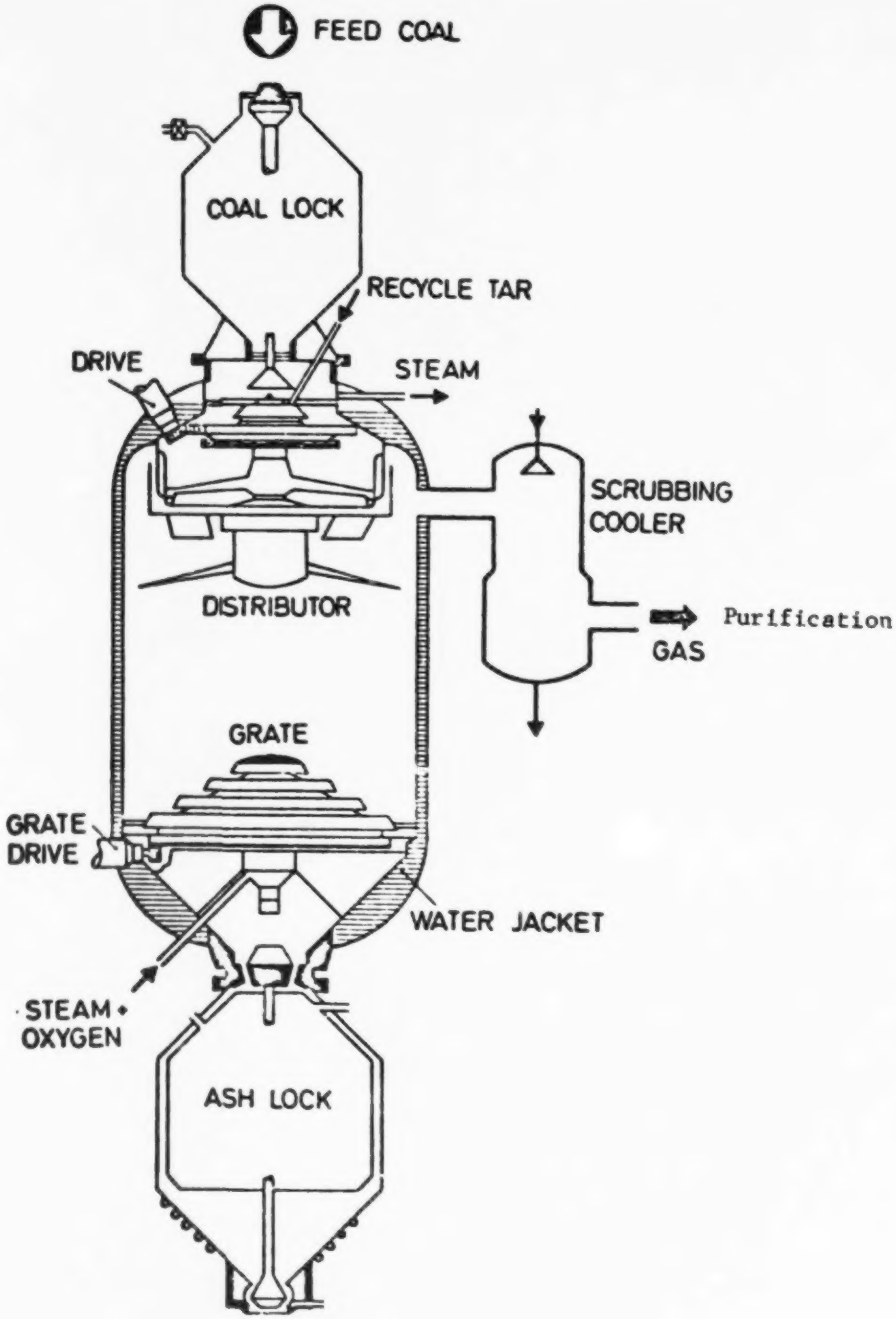

FIGURE 5.1.1. THE LURGI PROCESS (9)

TABLE 5.1.1. LURGI: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
300 x 10^6 SCF/day	Sasolburg, South Africa	South Africian Coal, Oil and Gas Corp.	Started operation in 1955 with 10 gasifiers; added 3 more in 1966. Synthesis gas for Fischer-Tropsch synthesis is major product. Gas cleanup with Rectisol; waste water cleanup with Phenosolvan. Output is limited by purification capacity not gasification (4).
55 x 10^6 SCF/day	Dorsten, Germany	Dorsten, Steinkolengas AG	Started operation in 1955. 6 gasifiers produces town gas from high volatile coal.
22 x 10^6 SCF/day	Melbourne, Australia	Morwell Gas and Fuel Corp.	Started operation in 1956. 6 gasifiers produces town gas from lignite.
4. x 10^6 SCF/day	Daud Khel, Pakistan	Pakistan Industrial Development Corp.	Started operation in 1957. 2 gasifiers produces synthesis gas from high volatile, high-sulfur coal.
25 x 10^6 SCF/day	Westfield, Scotland	The Scottish Gas Board	Started operation in 1960. 4 gasifiers produces town gas from weakly-caking high-volatile coal. In 1973-74 tests were carried out to determine the ability of a modified Lurgi gasifier to accept caking coals. These tests, sponsored by AGA and OCR, involved Illinois #5 and #6, Montana Rosebud Subbituminous, Pittsburgh Seam #8 and simulated run of the mine coal. All were successfully gasified. On basis of results, Lurgi will guarantee its gasifiers will accept Ill. #5 and #6 and the Montana Subbituminous. More work will be needed before that guarantee can be extended to other coals.
45 x 10^6 SCF/day	Coleshill, England	West Midlands Gas Board	Started operation in 1963. 2 gasifiers produces town gas from caking sub-bituminous coal.
15 x 10^6 SCF/day	Seoul, Korea	Naju, Honam Fertilizer Corp. Ltd	Started operation in 1963. 3 gasifiers. Produces synthesis gas from graphitc anthracite.
50 x 10^6 SCF/day	Pristina, Yugoslovia	Government	Started operation in 1972. 3 gasifiers. Produces town gas and synthesis gas from poor quality lignite. 3 more gasifiers are to be added. Use Rectisol for gas cleanup; Phenosolvan is used to cleanup waste water.

TABLE 5.1.1 (Continued)

Facilty	Location	Owner(s) or Contractor	Status/Operating History
160 x 10^6 SCF/day	Luenen, W. Germany	Steinkolen-Elektrizität AG (STEAG)	Started operation in 1972. 5 gasifiers (operated with air). Low-Btu gas (~180 Btu/SCF) is used to power a combined-cycle generating plant with a total net output of 170 MW. No provisions for desulfurization at present - to be added. Can be operated with slightly caking coals but coals with pronounced caking properties are not satisfactory. An 800 MW Unit of this type planned for operation in 1981. (7,8)
60 ton/hr Pilot	Pekin, Ill	Commonwealth Edison of Chicago	Estimate completion by 3rd quarter of 1976. Will involve 3 gasifiers. Low-Btu (150-200 Btu/SCF) fuel gas desulfurized with hot potassium carbonate) will be burned in modified existing boilers. Six different coals will be investigated during early tests.

REFERENCES:

(1) Hottel, H. C. and Howard J. B., New Energy Technology: Some Facts and Assessments, MIT Press, Cambridge, Mass. (1971).
(2) The Supply Technical Advisory Task Force-Synthetic Gas-Coal, Prepared by the Synthetic Gas-Coal Task Force for the Federal Power Commission (April 1973).
(3) "Evaluation of Coal Gasification Technology. Part II, Low- and Intermediate-Btu Gas", National Research Council, National Academy of Engineering, Washington, D.C. (1973).
(4) Hoogendoorn, Jan C., "Gas from Coal with Lurgi Gasification at SASOL", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 111-126, Chicago (1973).
(5) Anon,"Coal Gasification Plant Begins Operation Soon", Chemical and Engineering News (November 5, 1973).
(6) Squires, Arthur M., "Clean Fuels from Coal Gasification", p 340-346, Science Vol. 184, No. 4134 (April 19, 1974).
(7) Rudolph, Paul F. H., "A New Fossil-Fueled Power Plant Process Based on Lurgi Pressure Gasification of Coal", American Chemical Society (Division of Fuels Chemistry) Symposium on Coal Combustion in Present and Future Power Cycles, Volume 14, No. 2, pp 13-38 (May 1970).
(8) Krieb, K. H., "Combined Gas- and Steam-Turbine Process with Lurgi Coal Pressure Gasification", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 127-142, Chicago (1973).
(9) Moe, James M., "SNG from Coal via the Lurgi Gasification Process", ibid, pp 91-110.

5.1.2 Wellman-Galusha (McDowell Wellman Co.)

Description of the Process. A two-compartment feed bin is mounted on top of the gasifier. Disc valves permit fuel from the lower bin to flow continuously through the feed pipes to fill the fire chamber, and revolving grates discharge ash from below. The gasifier is water-jacketed and the inner wall is one inch steel plate, which requires no refractory lining.

Air (and/or oxygen) and steam enter from the bottom of the grate to gasify the coal. The standard Wellman-Galusha producer is shown schematically in Figure 5.1.2. An agitator producer is available which has a revolving horizontal arm, which also spirals vertically below the surface of the coal bed to retard channeling and maintain a more uniform fuel bed.

The product gas for the air-blown process has a heating value of 150-170 Btu/SCF and contains very little methane (less than 2.5 percent for bituminous coal). The gasifier can also be oxygen blown, in which case an intermediate-Btu gas would be produced.

The standard gasifier accepts only anthracite or coke; the agitator gasifier also accepts bituminous coal.

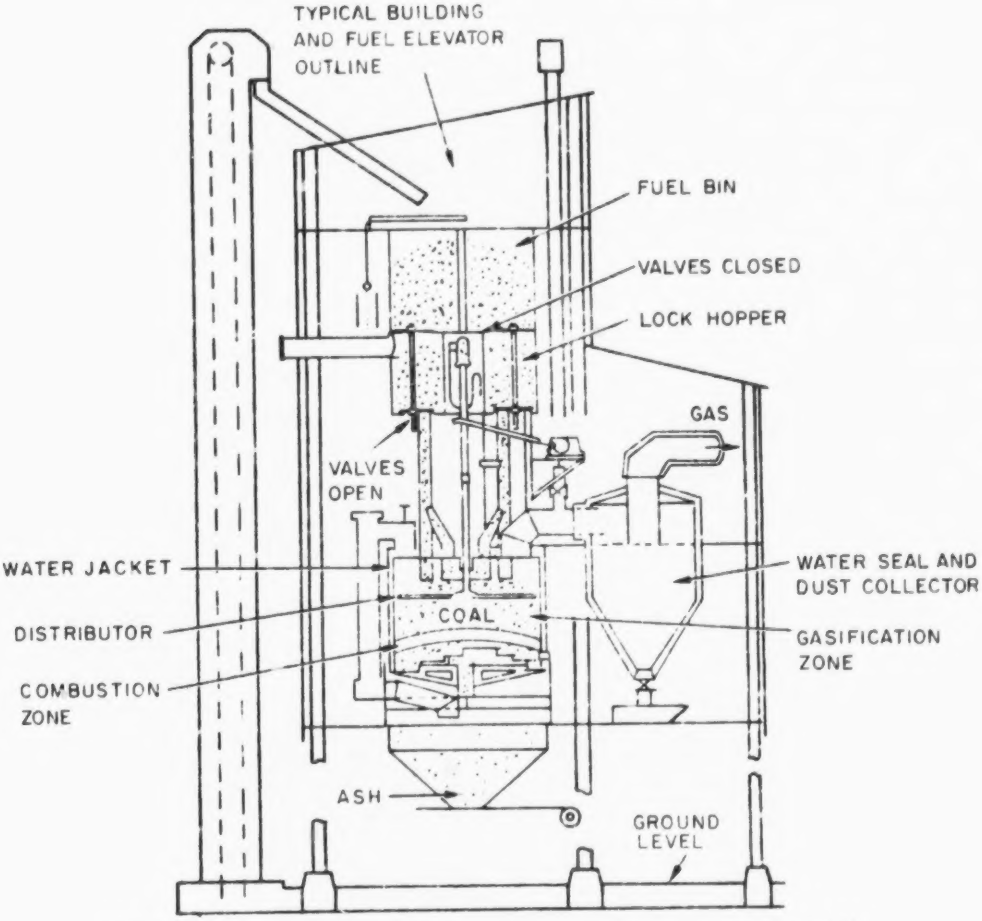

FIGURE 5.1.2. WELLMANN-GALUSHA FUEL GAS GENERATOR(1)

TABLE 5.1.2. WELLMAN-GALUSHA: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
			This is a commercial gasifier that has been in use in the U.S. for 30-40 years. Several are currently in operation including those at the following locations: National Lime and Stone, Carey, Ohio M.A. Hanna, Oregon Olin-Matheson, Kentucky New Jersey Zinc, Astabula, Ohio Glen-Gery Corp., Reading, Pa. There are also units operating in Canada, India, Tiwan and Cuba. Supplier in the U.S. is McDowell-Wellman Engineering Co., Cleveland, Ohio

(Contact: John F. Magnuson, McDowell-Wellman Engineering Co., Cleveland, Ohio (216) 621-9934)

References:

(1) "Gas Generator Research and Development: Survey and Evaluation" Phase One. Prepared by Bituminous Coal Research for the Office of Coal Research (August 1965).

(2) "Evaluation of Coal Gasification Technology. Part II. Low-and Intermediate- Btu Gas". Office of Coal Research, Washington (1973).

5.1.3 Bureau of Mines Stirred-Bed

Description of the Process. Many U.S. coals are unsuitable for use in moving-bed gasifiers because of their caking properties. The Bureau of Mines has studied the possibility of processing strongly caking coals in pressure gasifiers in which the fuel bed is continuously stirred. Vigorous, continuous stirring throughtout the bed breaks up coke formations and results in uniform bed conditions.

The unit shown schematically in Figure 5.1.3 has a diameter of 3.5 feet and is operated at moderate pressures. A variable-speed drive acting through two mechanisms rotates the shaft and moves it vertically in reciprocal motion. It does not appear necessary to extend stirring into the oxidation zone, and thus the lower rabble arm is not subjected to high temperatures in an oxidizing environment.

Air and steam are admitted below the grate. Temperatures are held below the ash fusion temperature to avoid forming clinkers which could clog the grate. Coal is fed intermittently through lock hoppers. Ash discharged by eccentric rotation of the grate is removed through lock hoppers.

The product gas has a heating value in the range 140 to 165 Btu/SCF.

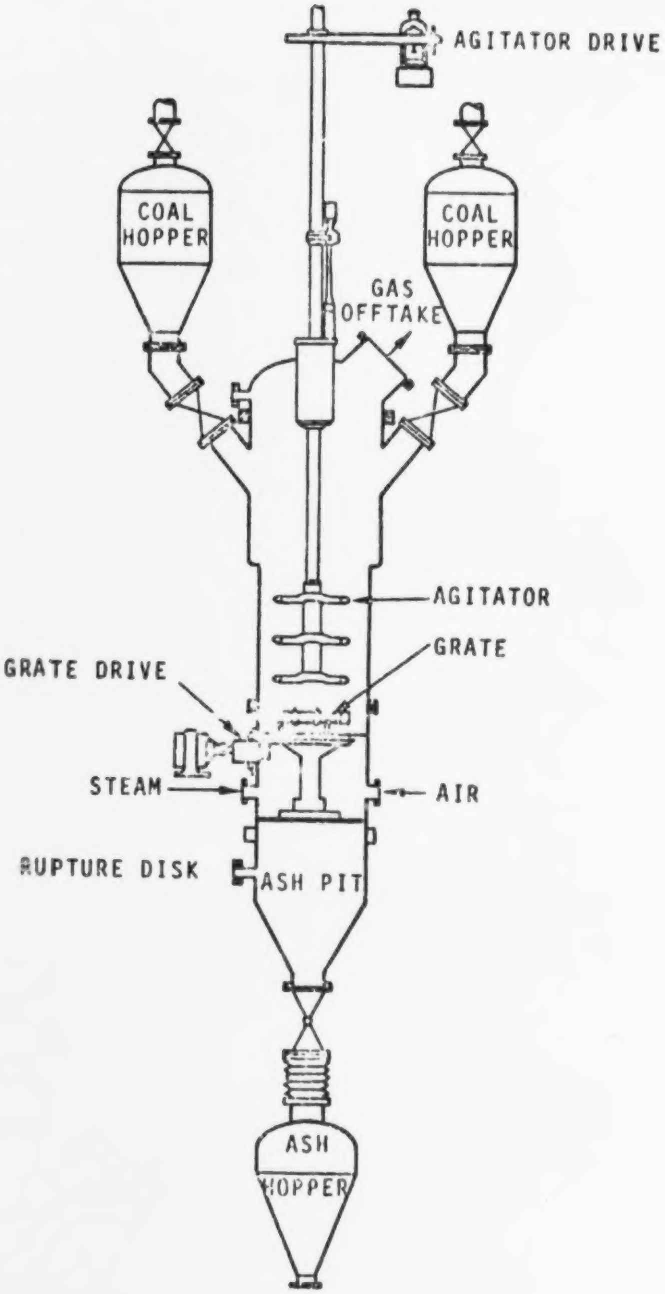

FIGURE 5.1.3. BUREAU OF MINES GASIFIER(1)

TABLE 5.1.3. BUREAU OF MINES STIRRED-BED PRODUCER: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
1200 lb/hr process development unit	Morgantown, W. Va.	U. S. Bureau of Mines	This unit has been in operation since 1972. Preliminary tests reported in 1972(1) demonstrated that strongly caking coal can be gasified in a stirred-bed producer. Presently studying optimization of operating conditions. Studies are to be carried out at pressures of 15, 100, 150, and 280 psi. Also, plan runs with TVA coal with the goal of providing conceptual design for commercial gasifiers for TVA. The OCR-TVA program calls for installation of 2 or more stirred-bed gasifiers at a TVA power plant. In addition, a process is to be developed for hot-gas cleanup.

(Contact: A. J. Liberatore, U. S. Bureau of Mines, Morgantown, West Virginia, (304) 599-7161)

(1) Lewis, P. S., Liberatore, A. J. and McGee, J. P., "Strongly Caking Coal Gasified by Stirred-Bed Producer", RI 7644, U. S. Bureau of Mines, Morgantown, West Virginia (1972).

(2) Rafuse, R. V., Goff, G. B., and Liberatore, A. J., "Noncaking Coal Gasified in a Stirred-Bed Producer", Bureau of Mines Clean Energy Program, Technical Progress Report, 77 (March 1974).

5.1.4. Gegas (General Electric Company)

Description of Process. General Electric is developing a moving-bed gasifier capable of producing low-Btu gas for combined-cycle power generating plants (Figure 5.1.4). Considerable emphasis has been given to the development of a coal-extrusion feed system in which fines and feed coal are compacted and injected into the gasifier in a single operation. Tar recovered from the off-gas is used as a binder. This unique feed system would permit the use of a wide variety of coals with the well-established moving-bed technology. A conventional lockhopper feed system is considered a development option.

The gasifier is air blown and will operate at a pressure of about 300 psi under dry ash conditions.

General Electric has devoted considerable research and development effort to a membrane purification system for use with the Gegas process.

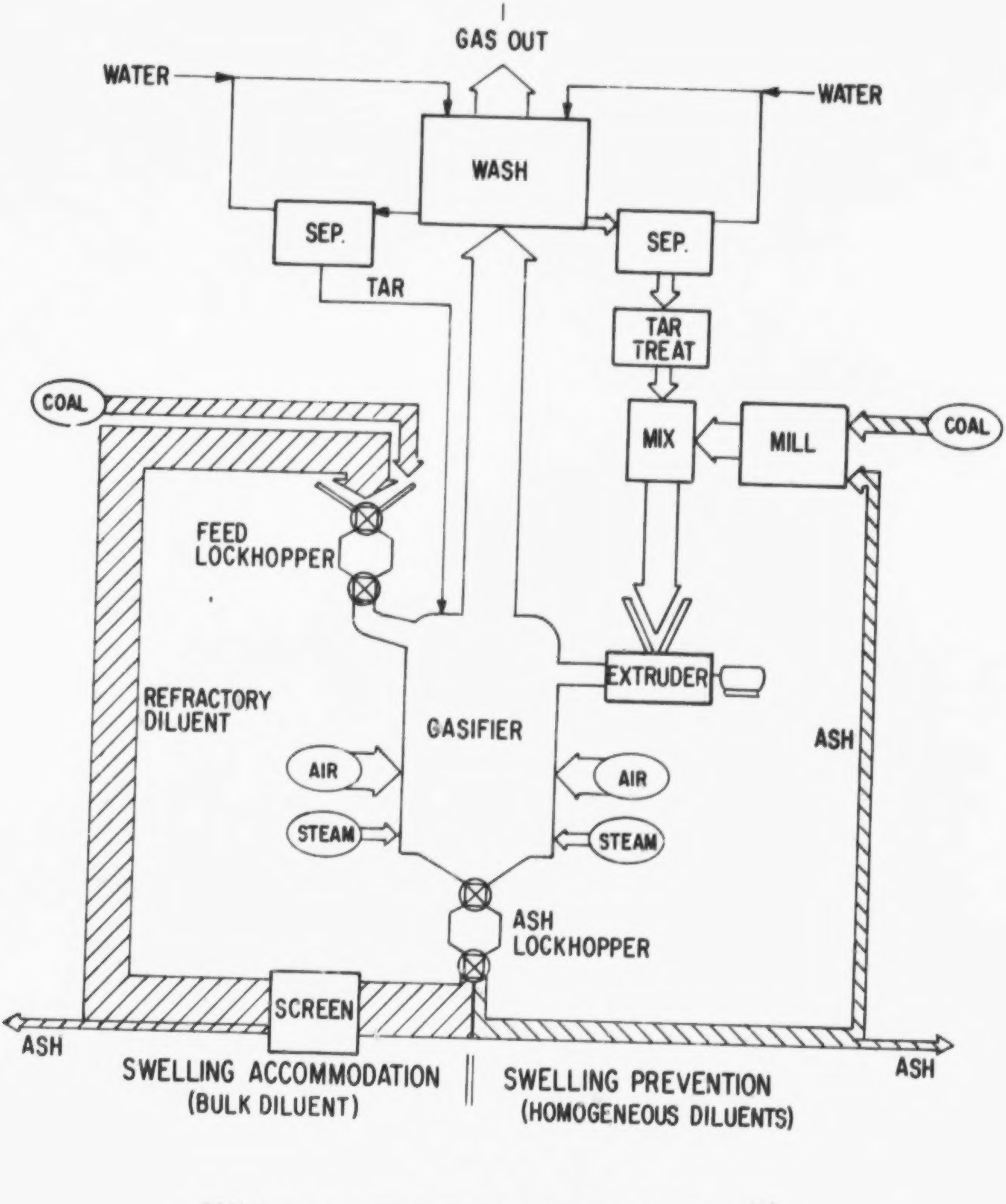

FIGURE 5.1.4. GEGAS PROCESS DEVELOPMENT OPTIONS(2)

TABLE 5.1.4. THE GEGAS PROCESS: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
50 lb/hr bench-scale gasifier	Schenectady, N.Y.	General Electric Co.	In operation since 1971. Considerable effort devoted to R&D of coal extrusion process for feeding coal to the gasifier, and on ability of process to accept a wide variety of coals
1200 lb/hr process development unit	"	"	Design stage.

Comments: Primary purpose is to produce low-Btu gas suitable for use in combined cycle power plants. GE is also doing considerable R&D on membranes for clean-up of the off-gas at 300 psi.

(Contact: Dr. Paul H. Kydd, General Electric Co., Schenectady, N.Y. (518-346-8771; X6535)).

References:

(1) Perry, Harry, "Coal Conversion Technology", Chemical Engineering, pp 95-96, Vol. 81, No. 15, (July 22, 1974).
(2) Courtesy General Electric Co., Schenectady, N.Y.

5.1.5 Kellogg Fixed-Bed Gasifier (MW Kellogg Co.)

A simplified flow diagram of the processes is shown in Figure 5.1.5. Sized coal with up to 20 percent fines is fed through lock hoppers to the gasifier which operates at about 15-25 psi. Steam and air, enriched air, or oxygen. are fed through the bottom of the grate.

Particulate matter is removed from the offgas leaving the gasifier by cyclones,and tars and other condensables are removed in a condenser with a steam drum.

The depth of ash in the gasifier is kept constant by use of a variable-speed revolving grate. Accumulated ash is dumped by gravity and removed from the gasifier by a lock hopper system. Stirring of the bed increases the variety of coals that can be accepted.

After desulfurization, the product gas has a heating value of about 150 Btu/SCF if the gasifier is air blown. If oxygen is used, the heating value is about 300 Btu/SCF.

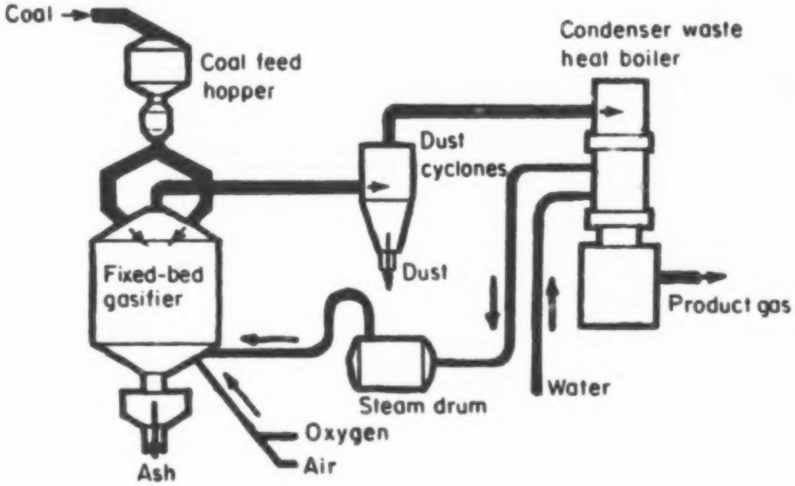

FIGURE 5.1.5. KELLOGG FIXED-BED GASIFIER

TABLE 5.1.5. KELLOGG FIXED-BED PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
4 ton/hr Pilot	Houston, Texas	M.W. Kellogg Co.	Will begin operation in mid-1975.
	(Contact: George Chedsey, M.W. Kellogg Co., Houston, Texas (713) 626-3236)		

Reference:

(1) Anon, Chemical and Engineering News, pp 17-18 (August 12, 1974).

5.2 Moving Bed/Slagging

In these gasifiers the moving bed is operated at temperatures that are high enough (2700 to 3000 F) to keep the ash in a molten state. Operation under slagging conditions results in increased steam decomposition and allows for higher throughput. It also results in the cracking of tars and oils so that a cleaner raw product is obtained.

A number of moving-bed/slagging gasifiers were built in Europe prior to 1950 and operated on a commercial scale. The Thyssen-Galoczy is representative of this type of gasifier.

5.2.1 Thyssen-Galoczy

Description of Process. The gasifier is blast-furnace shaped with three sets of tuyeres (Figure 5.2.1). Those comprising the lowest set are actually water-cooled burners in which gas from the process is burned in a mixture of steam and oxygen. Those at the upper levels admit oxygen. Only one of the two upper levels of tuyeres is used at any given time (the choice depends upon operating conditions). The gasifier operates at atmospheric pressure and 2900 F.

The base of the reactor has a refractory lining to resist erosion by molten slag. Coarse coal (1.5-2.5 inches) is charged to the gasifier by means of a bell arrangement. Slag is tapped at a level just below the gas burners, and iron, if any, is tapped at the bottom of the shaft.

The process was intended for use on any caking or noncaking fuel, and regardless of ash content or melting point. However, the only demonstrations appear to have been on closely sized coke of good quality. Under these circumstances the gas produced is composed principally of carbon monoxide (65-70 percent) and hydrogen (23-25 percent). Very little (0.2 percent) methane is produced.

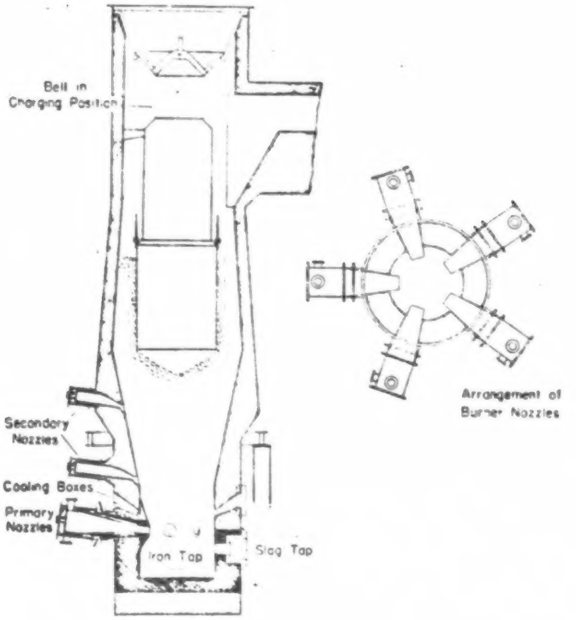

FIGURE 5.2.1. THYSSEN-GALOCSY SLAGGING GAS GENERATOR(1)

TABLE 5.2.1 THYSSEN-GALOCSY: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
2 ton/day Pilot	Duisburg-Hamborn, Germany	Thyssen'sche Gas und Wasserwerbe	Reliable information not available
40 ton/day Pilot	Wanne-Eickel, Germany	Krupp Treibstoffwerke	Tests were carried out in late 1943 and early 1944 with the hope of demonstrating that this unit could gasify any grade of fuel in any size or combination of sizes from No. 4 mesh to 3 inches, caking or noncaking and regardless of ash content. Testing was interrupted by the war and work was not resumed.(1)

References:

(1) Wright, C. C., Barclay, K. M., and Mitchell, R. F., Ind. Eng. Chem. pp 578-9, Vol. 40, (1948).

(2) Von Fredersdorff, C. C., Elliot, M.A., "Coal Gasification", In Chemistry of Coal Utilization, H. H. Lowry, Editor, Supplementary Volume, John Wiley and Sons, Inc., New York, pp 892-1022 (1963).

5.3 Fluid-Bed/Dry Ash

In fluidized-bed reactors, the gas flows through the bed and the size of the coal particles is such that the bed behaves like a fluid. In general, fluidized-bed reactors can accept fuels with wide variations in quality, and because of characteristically high heat transfer, bed temperatures are uniform and easily controlled. The very effective gas-solid contact leads to increased reaction rates and thus high volume efficiencies. On the other hand, pretreatment is required to prevent agglomeration, and carbon carryover in the product gas can result in high fuel losses, particularly with unreactive or friable fuels. Also, the range of possible operating conditions is restricted by the fluidization characteristics of the fuel.

5.3.1 Winkler (Davy Power Gas Company)

Description of the process. The Winkler process (Figure 5.3.1) is a commercially proven, fluidized-bed, atmospheric pressure system which can be either air-or oxygen-blown. Crushed coal is fed to the gasifier through variable speed screws. The gasifying medium fludizes the coal bed and gasifies the coal at uniform temperature. Oxygen (or air) and steam are added at both the bottom and the top of the bed. Top addition serves to increase the conversion of carbon in the gasifier.

About 70 percent of the ash is carried out of the generator with the product gas while the remainder leaves at the bottom of the fluid bed. To prevent the deposit of molten particles in the exit ducts, part of the waste heat recovery system is installed in the generator immediately above the gasification zone. This cools the gas sufficiently to prevent sintering of the fly ash on the walls.

The Winkler generator is applicable to a variety of coals; however, caking coals, as a rule, cannot be gasified without pretreatment which results in a lower overall conversion efficiency. The Davy-Power Gas Company (the American supplier) is studying the possibility of pressurizing the Winkler Generator.

The product gas from gasification with oxygen has a heating value of about 280 Btu/SCF, while that from air-gasification is about 120 Btu/SCF.

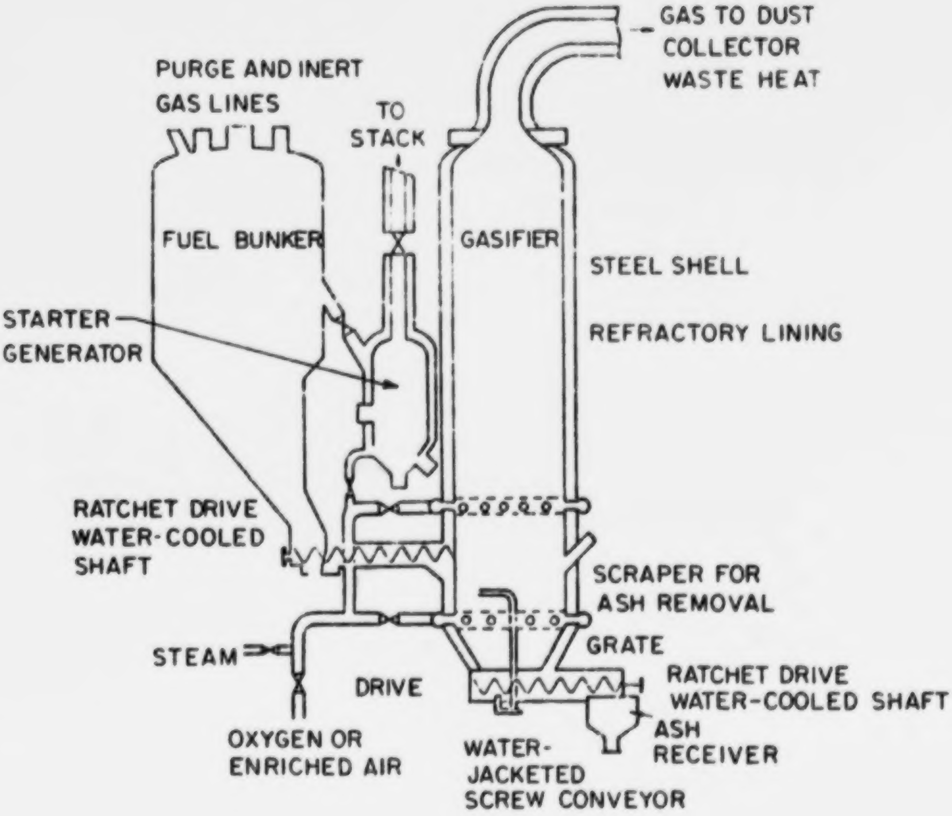

FIGURE 5.3.1. WINKLER GASIFIER (3)

TABLE 5.3.1 WINKLER: STATE OF THE ART[1]

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Commercial	Gorazde, Yugoslavia	Fabrika Azotnih Jendinjenja	Started operation in 1953. One gasifier with a normal operating capacity of 190 x 10^3 SCF/hr. Produces synthesis gas.
Commercial	Kutahya, Turkey	Azot Sanyyii TAS	Started operation in 1959. Two gasifiers, each with a normal operating capacity of 450 x 10^3 SCF/hr. Produces synthesis gas.
Commercial	Madras, India	Neyveli Lignite Corp	Started operation in 1959. Three gasifiers, each with a normal operating capacity of 1550 x 10^3 SCF/hr.

(Contact: I. N. Banchick, Davy-Power Gas, Lakeland, Florida (813) 646-7100)

References

(1) "Evaluation of Coal Gasification Technology, Part II Low--and intermediate--Btu Gas" National Research Council, National Academy of Engineering, Washington, D.C. (1973).

(2) Banchick, I.N., "The Winkler Process for Production of Low-Btu Gas from Coal", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 163-178, Chicago (1973).

(3) Indian Government Sponsored Study of Commercial Coal Gasification Processes, V. N. Kasturiranzan, M. Satyapal, R. R. Iyer, D. G. Rao and S. B. Chatterji, reproduced by Koppers Company, Inc., Pittsburgh (1973).

(4) Perry, Harry, "Coal Conversion Technology", Chemical Engineering, pp 88-102,(July 22, 1974).

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(1) A number of other commercial-scale Winkler gasifiers have been operated since 1926. This table, includes only those presently in operation (for a complete list see reference 2).

5.3.2 Snythane (Bureau of Mines)

Description of the Process. A flowsheet for the Synthane process is shown in Figure 5.3.2. Crushed coal is fed from a lock hopper system to a pretreater where the caking tendency of the coal is destroyed by treatment with oxygen and steam at 750 F. Pretreated coal, together with any volatile matter, and excess steam is fed into a two-zone gasifier which consists of a dense fluid bed in the top section and a dilute fluid bed in the bottom section.

Coal undergoes devolatilization and noncatalytic methanation at 1100 to 1470 F and 1000 psi in the dense fluid bed, and is then gasified with steam and oxygen in the dilute fluid bed at 1750 to 1800 F to produce the synthesis gas for the upper bed. The raw gas is quenched to remove tar, and H_2S is removed by scrubbing.

Operation with steam and oxygen produces a gas of about 300 Btu/scf while operation with steam and air yields a gas of about 180 Btu/scf. The process accepts all types of coal.

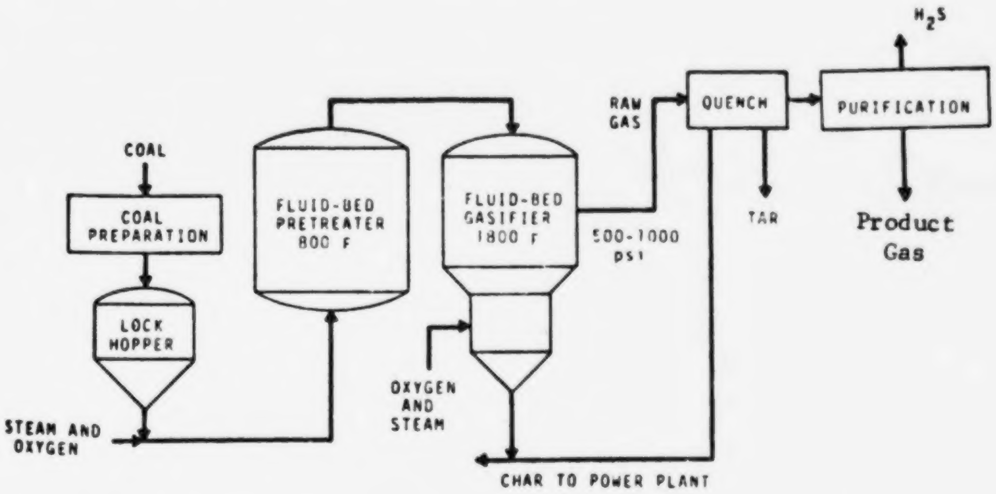


FIGURE 5.3.2. SYNTHANE PROCESS(4)

TABLE 5.3.2. SYNTHANE: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
75 ton/day Pilot	Bruceton, Pa.	U.S. Bureau of Mines	Construction about 60% completed as of July 1974. Expect completion in January 1975. Cost \$13 x 10^6. Studies on a wide variety of coals with a 40 lb/hr gasifier indicate process will accept any U.S. coal.
(Contact: J. Forney, U.S. Bureau of Mines, Pittsburgh, Pa., (412) 892-2400)			

References:

(1) Hottel, H. C., and Howard, J. B., New Energy Technology - Some Facts and Assessments, MIT Press, Cambridge Mass. (1971).

(2) "Evaluation of Coal Gasification Technology, Part II Low- and Intermediate- Btu Fuel Gas", R&D Report No. 74, Interim Report No. 1, Office of Coal Research, Washington, D.C. (1973).

(3) The Supply-Technical Advisory Task Force-Synthetic Gas-Coal, prepared by Synthetic Gas-Coal Task Force for the Federal Power Commission (April, 1973).

(4) Forney, A. J., Haynes, W. P., Elliot, J. J., Gasior, S. J., Johnson, G. E. and Starkey, J. D., Jr., "The Synthane Coal-to-Gas Process", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp. 199-208, Chicago, (1973).

5.3.3 CO_2 Acceptor (Consolidation Coal Co.)

Description of the Process. There are two reactors, the gasifier and the regenerator (Figure 5.3.3). Crushed coal is fed through lock hoppers to the gasifier where it is devolatilized and then gasified at 150 psi and at temperatures in the range of 1500 to 1550 F. Introduction of the feed at the bottom of the reactor fluidized bed provides sufficient residence time for the cracking of coal volatiles.

The devolatilized char is fluidized and partially gasified by steam. Heat for devolatilization and gasification is provided by reaction of carbon dioxide in the gas stream with the acceptor, calcined dolomite. Hot calcined acceptor flows by gravity from the regenerator to the gasifier and is introduced above the gasifier char bed. Acceptor showers through the char bed supplying both sensible and chemical heat and collects in a reduced cross-section boot at the bottom of the gasifier.

Spent acceptor is segregated from the lower density char and carried pneumatically to the regenerator where it is calcined at 1870 F. Heat for this process is provided by combustion of residual char with air. The product is an intermediate-Btu gas.

The process is designed to operate with lignite or subbituminous coal.

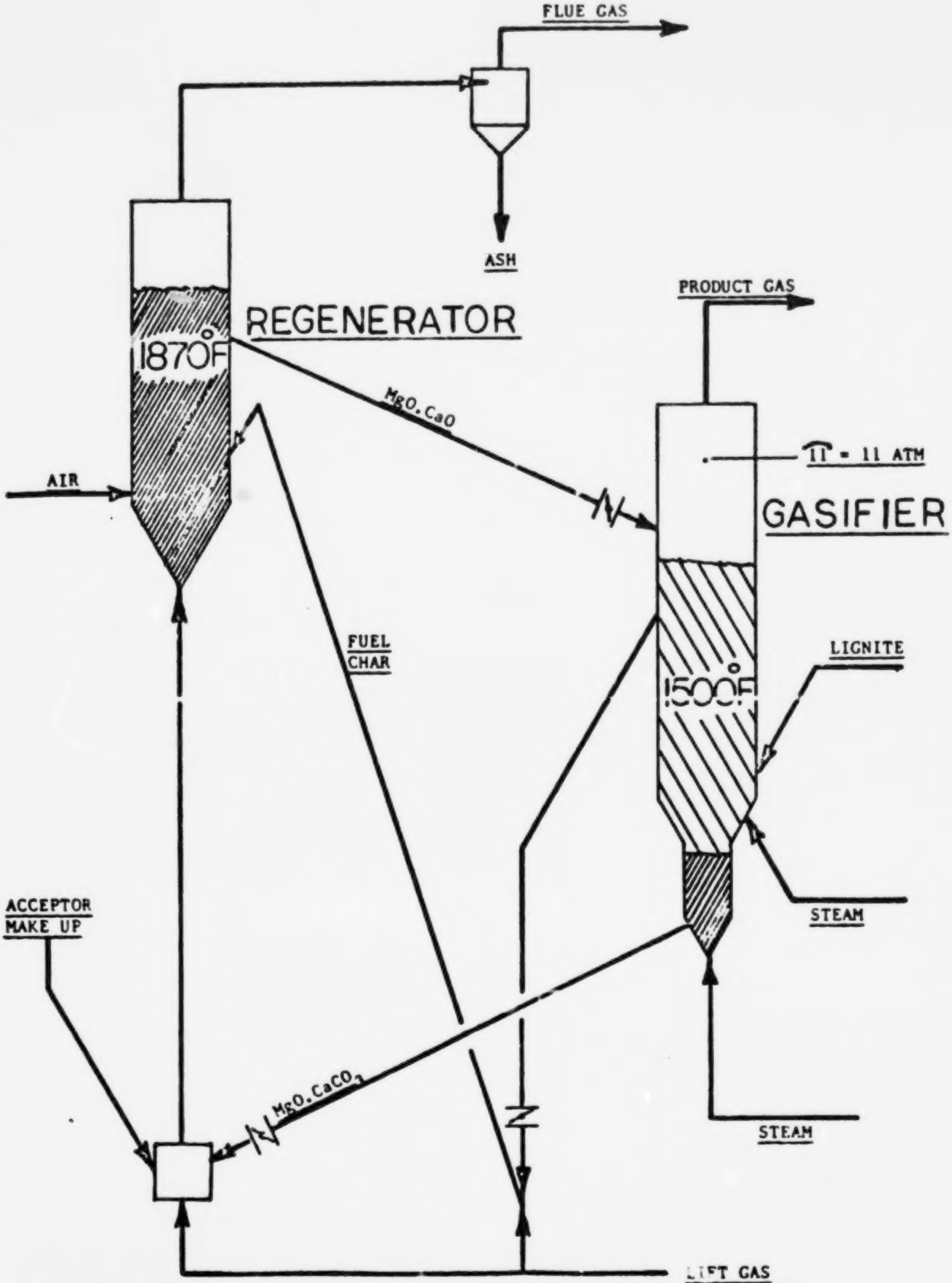


FIGURE 5.3.3. CO_2 ACCEPTOR PROCESS DIAGRAM(4)

TABLE 5.3.3. CO_2 ACCEPTOR: STATE OF THE ART

Facility	Location	Owner(s) or Contractor	Status/Operating History
40 ton/day Pilot plant	Rapid City, S.D.	Consolidation Coal Co., (OCR and AGA support) (plant constructed and operated by Stearns Roger Corp.)	- Plant cost about $\$9.3$ x 10^6. In operation since April 1972. - 25 runs were completed in the period 4/72 - 5/74; three were of 100 or more hours. - Mechanical problems in earlier runs slowed the acquisition of fundamental data. More recent runs have been more successful. - Problem of refractory failures has been solved. - Corrosion of fired heaters by H_2S has been eliminated by use of ZnO system to remove H_2S. However, formation of metal carbides by carbon deposition still resulted in loss of metal. Hope to solve this by adding steam to the gas going to these burners. - Problems with char combustion and plugging of acceptor lines during start-up also appear to be solved. - Designed for use with lignite and subbituminous coal. - Designed to operate at pressure of 150 to 300 psi and temperatures up to 1800 F.
	(Contact: Carl E. Fink, Consolidation Coal Co., Rapid City, S.D., (605) 342-6416)		

References:

(1) Hottel, H. C., and Howard, J. B., New Energy Technology - Some Facts and Assessments, MIT Press, Cambridge, Mass., (1971).

(2) "Evaluation of Coal Gasification Technology, Part II, Low- and Intermediate-Btu Fuel Gas, Office of Coal Research, Washington, D.C. (1973).

(3) The Supply-Technical Advisory Task Force-Synthetic Gas-Coal, prepared by Synthetic Gas-Coal Task Force for the Federal Power Commission (April, 1973).

(4) Fink, Carl E., "The CO_2 Acceptor Process", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 301-310, Chicago (1973).

(5) Annual Report for Calendar Year 1972, Office of Coal Research, U.S. Dept. of Interior, Washington (1973).

(6) Annual Report 1973-74, Office of Coal Research, U.S. Dept. of Interior, Washington (1974).

5.3.4 Exxon

Description of the Process. Exxon is developing a fluid-bed gasifier for production of intermediate-Btu gas from coal. Very little has been published about this process, though it is known that it operates at pressures of a few hundred psi and does not require oxygen. Presumably, the heat required for gasification is obtained from hot char produced in another reactor by partial combustion of coal with air. Coal is fed to the gasifier by a lock hopper system.

TABLE 5.3.4 EXXON: STATE OF THE ART

Facility	Location	Owner(s) or Contractor	Status/Operating History
.5- ton/day process development unit	Baytown, Texas	Exxon Oil	In operation since 1967. Results demonstrate process wroks with wide variety of coals.
500 ton/day pilot	" "	" "	Construction is expected to be completed by fall of 1976. (Contract has been let.)

Comments: Proprietary process. Very little information available.

(Contact: R. Pennington, Exxon Corp., Baytown, Texas (713) 427-5711)

References:

(1) Anon, "New Processes Brighten Prospects of Synthetic Fuels from Coal", p. 97, (April, 1974).

5.3.5 HRI Gasification (Hydrocarbon Research Incorporated)

The HRI fluid-bed process is shown schematically in Figure 5.3.5. Coal fines are charged to a hopper which is then pressurized to transfer the coal to the gasifier which operates at 400 psi. Steam and oxygen (or air) are preheated to about 1000 F before entering the gasifier.

Coal flows downward through the gasifier and dry ash is removed from the bottom. The gas is scrubbed to remove particulates and desulfurized.

If the gasifier is air blown, a 150 Btu/SCF fuel gas is obtained. Gasification with oxygen increase the heating value to about 250-350 Btu/SCF.

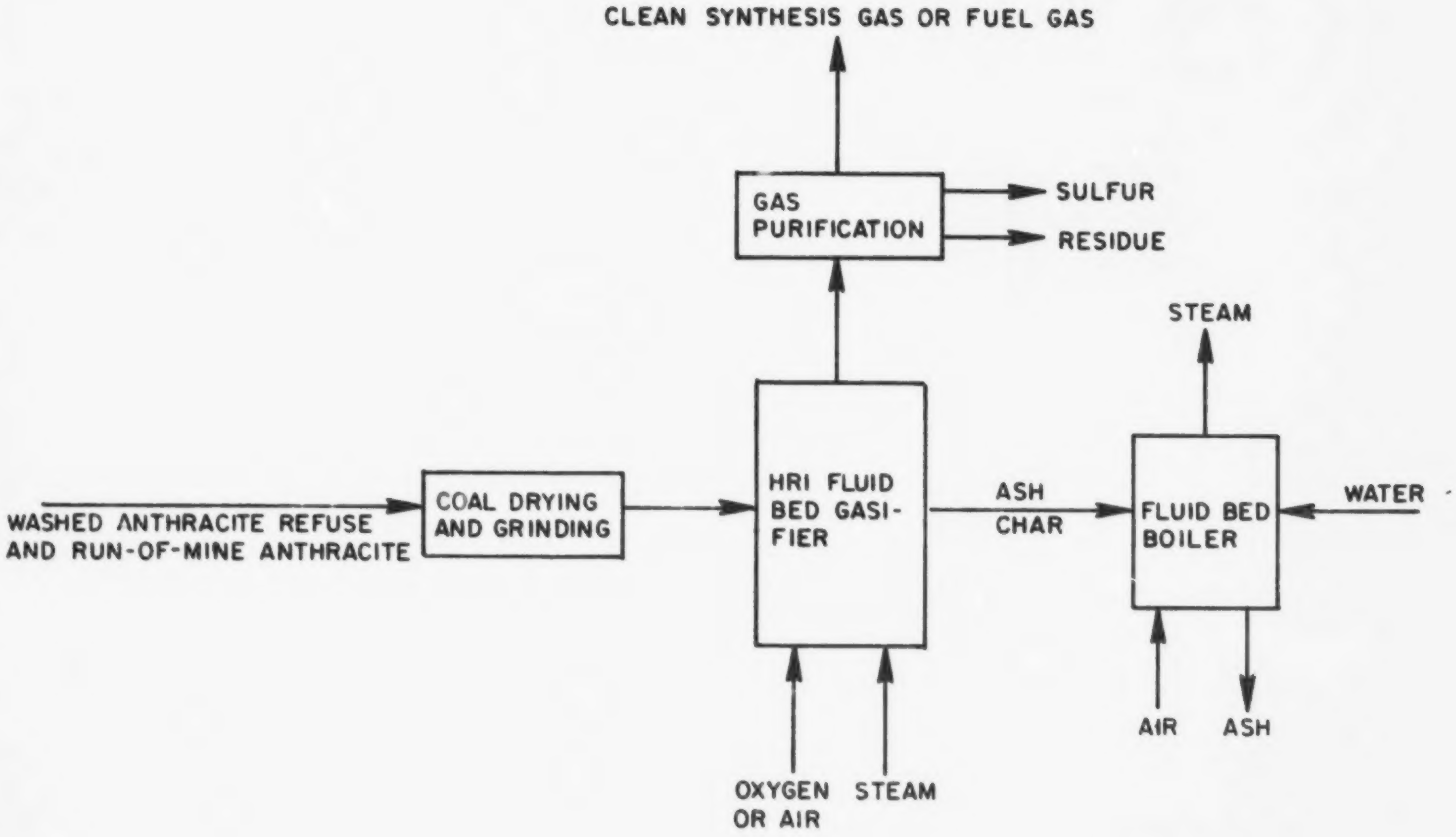


FIGURE 5.3.5. SCHEMATIC FLOW SHEET OF ANTHRACITE GASIFICATION PILOT PLANT

TABLE 5.3.5. HRI: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
10 ton/day	Trendton, N. J.	Hydrocarbon Research, Inc.	In operation for a period of about three months in 1958. Six runs were made, the last one for a 2 week duration. Run was terminated voluntarily and inspection revealed unit was in good condition. In 1972 the gasifier was modified to gasify bituminous coal under contract with Bureau of Mines in conjunction with its synthane program. That work is completed. (Original unit was designed primarily to gasify anthracite refuse and/or run-of-mine anthracite.)

(Contact: Mr. J. Y. [illegible]ingston, Hydrocarbon Research, Inc., N.Y. (212) 349-1480

Reference:

(1) "Gasification of Anthracite Breaker Refuse and Anthracite to Produce Clean Fuels", A Proposal from HRI to OCR (1972).

5.3.6. COGAS (Cogas Development Company)

Description of Process. COGAS is a fluidized-bed process that produces both synthetic crude oil and gaseous products from coal. The initial step involves a multi-stage pyrolysis of the coal to extract the volatile matter and produce a low-volatile char. This part of the process together with fixed-bed hydrotreating of the raw liquid from pyrolysis is the COED process described in Chapter 7. It is the source of synthetic crude.

The char that is produced is gasified to produce an intermediate-Btu gas suitable for upgrading to SNG. Because the gasification process is proprietary in nature, little information has been published. The char is gasified with steam or steam and hydrogen at 1600 to 1700 F and low pressures (15 to 40 psi). The heat necessary for gasification is provided by combustion of a portion of the char in air "with provision for preventing the air from mixing with the product gas".

Part of the raw gas from the gasifier is used in the hydrotreater to upgrade the raw oil produced by pyrolysis. Gaseous products from pyrolysis are combined with the raw gas from the gasifier and purified to produce a product with a heating value of about 400 Btu/SCF.

TABLE 5.3.6. COGAS: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
100 ton/day Pilot	Leatherhead, England	Operated for Cogas Development Co. by the British Coal Utilization Board	Operational since March 1974. Mechanical problems were few and were overcome with relative ease. Past several months have been devoted to proving process operability.
5 ton/day Pilot	Plainsboro, N.J.	Cogas Development Co. (a consortium of FMC Corp., Consolidated Natural Gas, Panhandle Eastern Pipe Line, Republic Steel, Rocky Mountain Energy and Tenn. Gas Transmission Co.)	Operational since late spring of 1974. No information available. Through calendar 1974, Cogas Development. This included bench scale work, process and economic studies, and the design, construction and operation of two pilot plant facilities.

(Contact: Howard Malakoff (General Manager), Cogas Development Co., Princeton, N. J. (609) 452-2300

References:

(1) Dierdorff, L. H., Jr., and Bloom, R., Jr., "The COGAS Project - One Method of Coal to Gas Conversion", paper presented at the West Coast Meeting of the Society of Automotive Engineers, Portland, Oregon (August 20-23, 1973).
(2) Perry, Harry, "Coal Conversion Technology", Chemical Engineering, pp 88-102 (July 22, 1974).

5.3.7. Bituminous Coal Research

On April 4, 1974, OCR announced that a 50-month contract had been awarded to BCR to develop a multiple fluidized-bed gasification process to produce low-Btu and intermediate-Btu fuel gases from coal.

TABLE 5.3.7. BITUMINOUS COAL RESEARCH

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Bench-scale	Monroeville, Pa.	Bituminous Coal Research (funded by OCR)	A $2,575,000, 50-month contract was awarded in April 1974. This will cover bench-scale and process development unit work that will be the basis for design of a pilot plant. Some laboratory investigations have already been carried out in a small fluidized-bed batch reactor to verify proposed rate equations, to determine the degree of steam or CO_2 decomposition that could be achieved in a reactor of reasonable size, and to provide physical data such as minimum fluidizing velocity, etc.

References:
(1) Department of the Interior News Release, April 4, 1974.
(2) Annual Report 1973-74, Office of Coal Research, U.S. Dept. of Interior, Washington, D.C. (1974).

5.4 Fluid Bed/Agglomerating Ash

Selective, continuous removal of ash from a fluidized bed rich in carbon can be a difficult problem. One solution is to permit ash particles to reach temperatures at which agglomeration occurs. When they become sufficiently heavy, ash agglomerates separate from the bed and can be removed.

The Union-Carbide-Battelle process to be described in this section also makes use of ash agglomerates to supply heat to the gasifier and by so doing eliminates the need for a costly oxygen plant when intermediate-Btu gas is to be produced.

5.4.1 U-Gas (Institute of Gas Technology)

Description of the process. Caking coals are pretreated with air at 800 F and 350 psia to render them noncaking (Figure 5.4.1). Treated coal overflows into the fluidized-bed gasifier where it reacts with air and steam at about 1900 F. Part of the fluidizing gas enters through a grid which is sloped toward one or more cones contained in the grid. The rest flows upward at high velocity through the throat at the cone apex creating a submerged jet within the cone. The temperatures generated within the jet are somewhat higher than in the rest of the bed. As carbon is gasified in and near the jet, ash is heated to its softening point, the sticky ash surfaces cling to one another, and ash agglomerates grow in the violently agitated jet. When they become heavy enough, the agglomerates fall counter to the high-velocity gas and are separated from the bed.

Gas above the bed is at a temperature of between 1500 and 1900 F and the residence time is sufficient to allow for thermal cracking of tars and oils. Most of the dust in the raw gas is removed by internal cyclone separators and returned to the fluidized bed. After separation of fine dust in external cyclones, the gas is at 1550 F. IGT is developing a high temperature Meissner (800 F) process for desulfurization of this gas.

Gasification with air and steam produces a gas with a heating value of about 150 Btu/scf. When oxygen is used for gasification the heating value is about 300 Btu/scf.

The process can accept all ranks of coal and lignite.

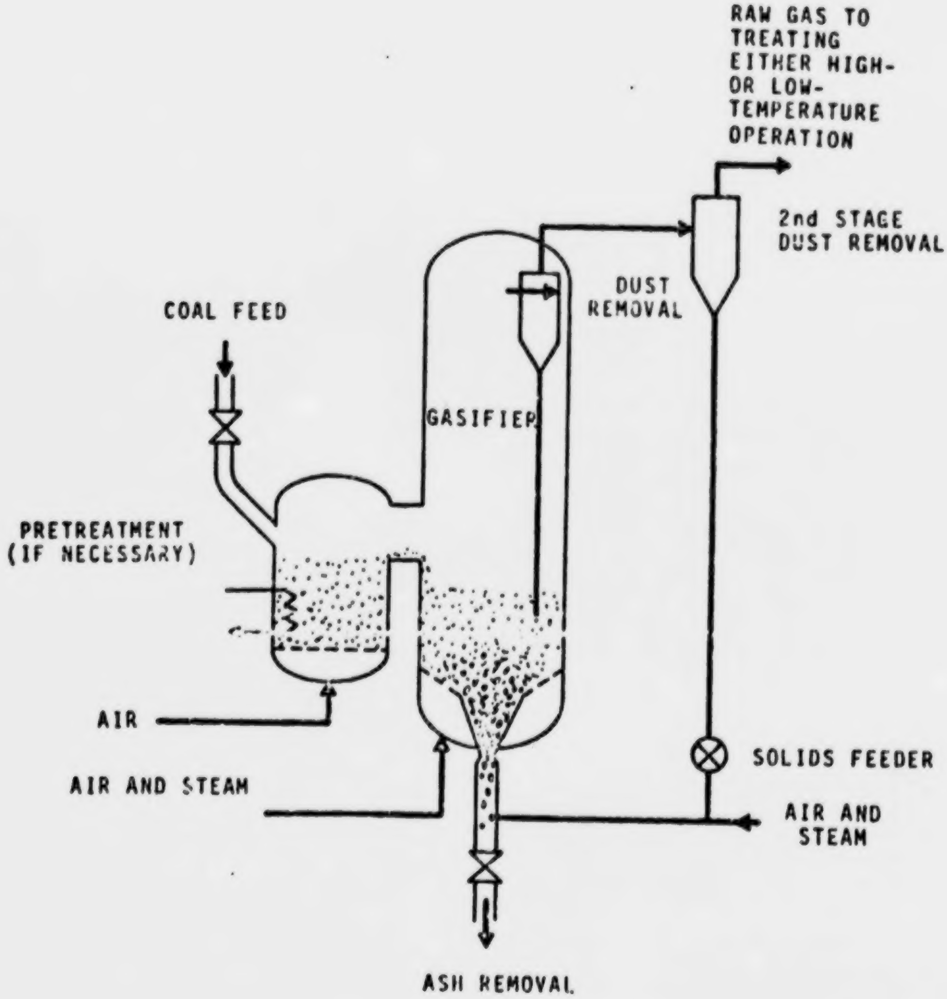


FIGURE 5.4.1. GASIFIER TO BE USED IN IGT'S U-GAS SYSTEM(1)

TABLE 5.4.1 U-GAS: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
10 ton/hr Pilot	n.a.	IGT	Design stage. Seeking funding. Presently, studies are being made with a 4 ft diameter agglomerating-ash gasifier. In operation since June 1974.

(Contact: John W. Loeding, IGT, Chicago, (312) 225-9600, Ext. 841)

References:

(1) Loeding, John W., and Tsaros, Constantine, L., "IGT U-Gas Process", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 241-273, Chicago (1973).

(2) "Evaluation of Coal-Gasification Technology, Part II, Low-and Intermediate-Btu Fuel Gases", National Research Council, National Academy of Engineering, Washington, D.C. (1973).

(3) Perry, Harry, "Coal Conversion Technology", Chemical Engineering, pp 88-102,(July 22, 1974).

5.4.2. Westinghouse

Description of the Process. This process, which is in the design stage, is intended to operate in conjunction with a combined cycle power plant. The gasification process is shown schematically in Figure 5.4.2. Crushed coal is dried in a fluidized bed and transported to the devolatilizer-desulfurizer unit. Here devolatization, desulfurization, and hydro-gasification are combined in a single, recirculating fluidized-bed reactor operating at 1300 to 1700 F and 20 to 30 atmospheres.

Dried coal fed into a central draft tube of this reactor is diluted with large quantities of recycled solids--char and lime sorbent. These solids, flowing at rates up to 100 times the coal feed rate, prevent or control the agglomeration of the coal feed as it devolatilizes.

A dense dry char is collected in the fluidized bed at the top of the draft tube. Lime is added to this bed to remove sulfur. Spent sorbent is withdrawn from the reactor after stripping out the char. The final gasification of the low-sulfur char is conducted in a fluidized bed with a lower leg which serves as a combustor. In this section, char from the devolatilizer-desulfurizer is burned with air at about 2100 F to provide the gasification heat. Ash agglomerates fall to the lower bed leg are removed.

In the upper section of the bed gasification occurs at 1800-2000 F. After removing particulates with cyclone separators, the clean fuel gas passes to the gas turbine plant.

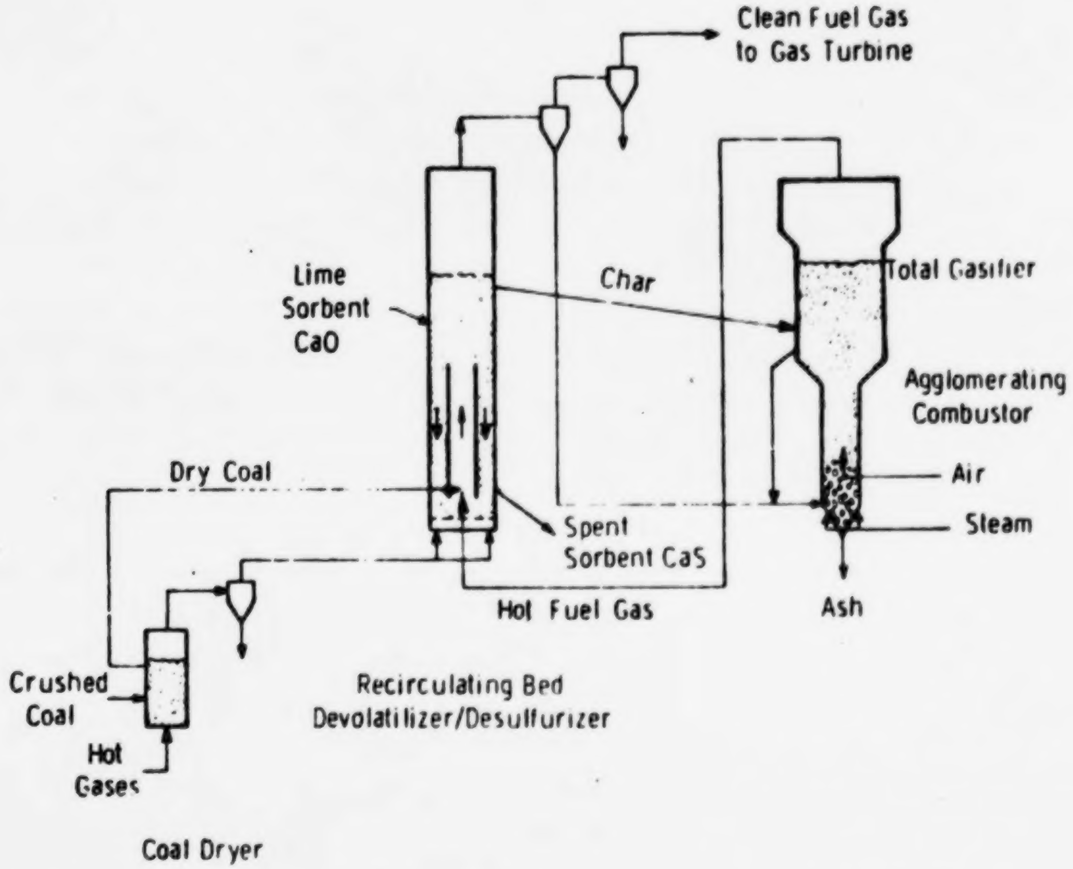


FIGURE 5.4.2. WESTINGHOUSE MULTISTAGE FLUIDIZED BED GASIFICATION PROCESS(4)

TABLE 5.4.2 WESTINGHOUSE: STATE OF THE ART

Facility	Location	Owner(s) or Contractor	Status/Operating History
1200 lb/hr process development unit	Waltz Mill, Pennsylvania	Westinghouse (supported by OCR, Public Service Indiana, AMAX Coal Company, Bechtel Corp., and Peabody Coal Co.)	Unit, which cost $5.5 x 10^6 for design and construction, is now mechanically complete and in the pre-commission stage. Expect first hot tests in January 1975. Unit includes gasifier and devolatizer/desulfurizer. Test program will establish inherent operating characteristics of the gasification system and provide data for design of larger units. If significant success is achieved with this unit, a decision could be made to ge directly to design of a full-size generating pilot plant gasifier to be used with a combined-cycle plant of about 120 MW. This is expected to be located at Dresser Station, Terre Haute, Indiana.(1)

(Contact: John Holmgren, Westinghouse, Waltz Mill, Pennsylvania, (412) 722-5552).

References

(1) "Coal Technology: Key to Clean Energy", Annual Report 1973-74, Office of Coal Research, Washington, D.C. (1974).

(2) Archer, D.H., Vidt, E.J., Keairns, D.L., Morris, J.P., and Chen, J.L.,"Coal Gasification for Clean Power Production", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 447-484, Chicago, (1973).

(3) Evaluation of Coal Gas Technology, Part II, Low-and Intermediate-Btu Fuel Gases, National Research Council, National Academy of Engineering.

(4) Archer, D. H., Kearns, D. L., and Vidt, E. J., "Development of a Fluidized-Bed Coal Gasification Process for Electric Power Generation", presented at the 4th Synthetic Fuels from Coal Conference, Oklahoma State University, Stillwater, Oklahoma (May 6-7, 1974).

5.4.3 Ash Agglomeration (Union Carbide/Battelle)

Description of Process. A simplified flow sheet for this process is shown in Figure 5.4.3. Crushed, dried coal is charged to the tapered transition zone of the gasifier which operates at 1800 F and 100 psi. Superheated steam enters below the fluidized-bed distribution plate. Hot ash agglomerates, flowing countercurrently, provide heat to support the gasification reactions.

Coal is converted to gas and char as it moves upward through the gasifier. Char separated from the raw exit gas is sent to the burner where it is combusted with air to reheat ash agglomerates. Ash-free flue gas from the burner passes through a heat recovery system and is expanded in a gas turbine to generate process power.

Ash agglomerates, stripped of carbon by the upward flow of steam in the gasifier, leave the bottom of the gasifier and are recycled to the burner.

The raw product gas is cleaned by cyclones and Venturi scrubber systems.

Because the coal fed to the gasifier is greatly diluted by a large quantity of circulating ash, the process is expected to accept caking coals without pretreatment. Also, the generation of hot ash agglomerates in a vessel separate from the gasifiers permits production of an intermediate-Btu gas without the usual requirement of an oxygen plant.

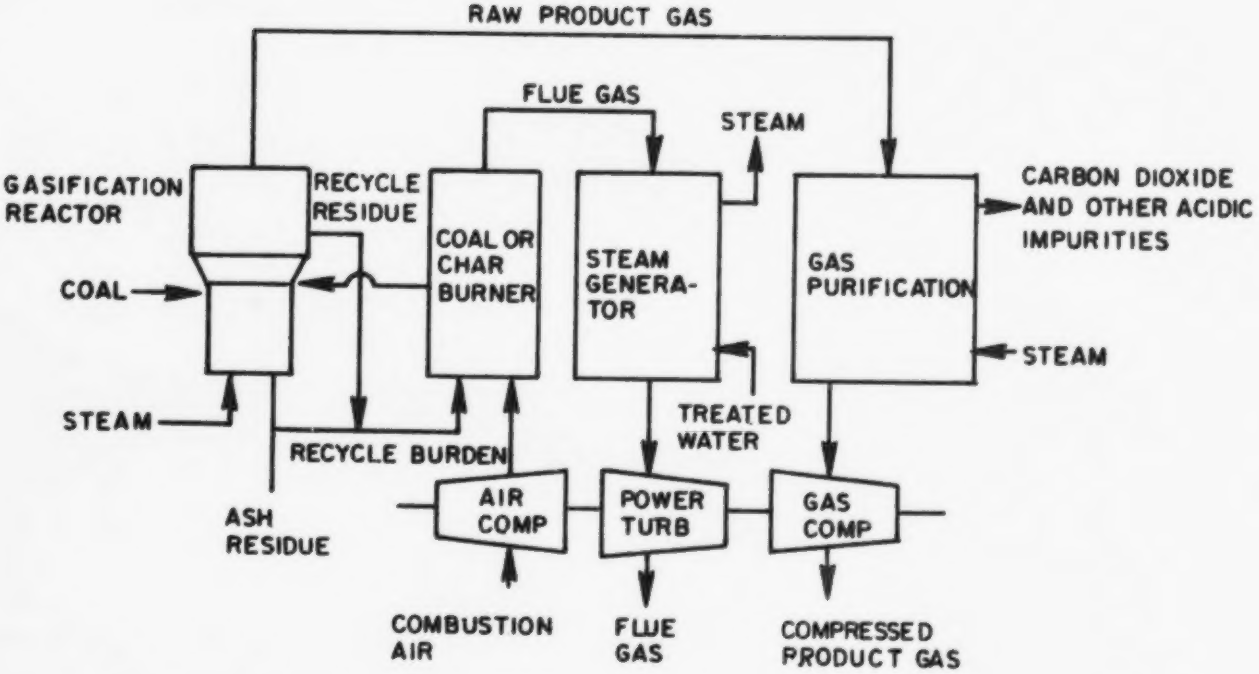


FIGURE 5.4.3. UNION CARBIDES' AGGLOMERATED ASH PROCESS (3)

TABLE 5.4.3. ASH AGGLOMERATION

Facilty	Location	Owner(s) or Contractor	Status/Operating History
1200 lb/hr process development unit	West Jefferson, O	Battelle (Funded by OCR and AGA) (Patent for the process is held by Union Carbide.)	Under construction. Estimate completion in first quarter of 1975. Unit will not include a gas purification system. Earlier studies included bench-scale studies of both the coal burner and the gasifier. Erosion by fly ash from the burner at simulated turbine conditions was also investigated.

(Contact: W. M. Goldberger, Battelle Columbus Labs., (614) 299-3151)

References:

(1) Corder, W. C., Batchelder, H. R., and W. M. Goldberger, "The Union Carbide/Battelle Coal Gasification Process Development Unit Design", Presented at the Fifth Synthetic Pipeline Gas Symposium, Chicago (Oct. 1973).
(2) "Evaluation of Coal-Gasification Technology, Part II, Low- and Intermediate- Btu Fuel Gas", Report by National Research Council, National Academy of Engineering.
(3) Goodridge, E., "Status Report: The AGA/OCR Coal Gasification Program", Coal Age 78, 54-59 (January 1973).

5.5 Entrainment Reactors

In entrainment or suspended bed reactors, pulverized coal is carried along with the gas. The major advantage of this type of system is the ability to accept all types of coal. Particles undergo only occasional collisions and, therefore, caking tendencies are of no consequence. Carbon carry-over is usually high and separation of ash solids from gases is a problem. With cocurrent flow, the temperature of the exist gas is high and a heat recovery system is required if good thermal efficiency is to be achieved. Overall energy production rates per unit volume of space in entrained gasifiers are much greater than those for moving-bed and fluid-bed gasifiers because of high reaction rates resulting from the large surface areas of the fuel particles.

5.5.1 Bigas (Bituminous Coal Research Inc.)

Description of the Process. A flow diagram for the Bigas process is shown in Figure 5.5.1. Piston feeders charge dried pulverized coal and steam into the two-stage gasifier near the bottom of stage II (upper section) where it mixes with hot gas rising from stage I and is devolatilized and partially gasified. Residual char entrained in the raw product gas leaving the top of the gasifier is separated by cyclones and recycled to Stage I where it is gasified with steam and oxygen under slagging conditions (2700-2800 F). Raw gas from the cyclones passes through a water quench system and a desulfurization unit.

If oxygen and steam are used for gasification, the product gas has a heating value of 380 Btu/SCF, with air and steam the heating value of the gas is 210 Btu/SCF.

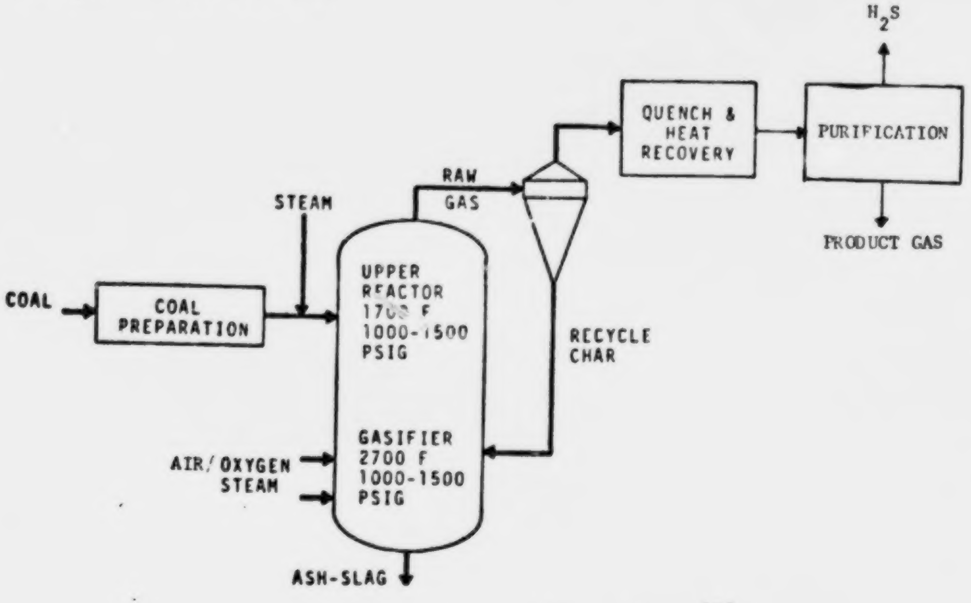


FIGURE 5.5.1. BIGAS PROCESS(3)

TABLE 5.5.1. BIGAS: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
5 ton/hr pilot	Homer City, Pa.	Bituminous Coal Research (with support from OCR and AGA) (Stearns Roger Corp. is responsible for construction)	Under construction. Estimate completion in early 1975. Estimated cost is about $\$25 \times 10^6$. This will be a fully integrated plant. Lab- and process development-scale (6000 SCF/hr) studies of methanation are being carried out to determine optimum operating conditions for the pilot plant. Acid gas removal by Selexol process.

(Contact: Robert Grace, Bituminous Coal Research, Pittsburgh, Pa., (412) 327-1600)

References:
(1) Hottel, H. C. and Howard, J. B., New Energy Technology: Some Facts and Assessments, MIT Press, Cambridge, Mass (1971).
(2) "Evaluation of Coal Gasification Technology. Part II, Low- and Intermediate-Btu Fuel Gases; National Research Council, National Academy of Sciences, Washington, D.C. (1973).
(3) Grace, Robert J., "Development of the Bigas Process", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 179-198, Chicago (1973).
(4) "Clean Energy from Coal - A National Priority", 1973 Annual Report, Office of Coal Research, Washington, D.C. (1973).
(5) "Coal Technology: Key to Clean Energy", Annual Report 1973-74, Office of Coal Research, Washington, D.C. (1974).

5.5.2 Combustion Engineering Inc.

Description of Process. Combustion Engineering has undertaken a four-phase program for the development of a process for conversion of coal to a clean fuel gas for electric power generation. An entrainment-type gasifier operating at atmospheric pressure will generate low-Btu gas for use in either a conventional steam or combined-cycle power plant.

The combustion and reducing chambers of the gasifier are enclosed by water-cooled walls that are studded and refractory-covered. Coal and recycled char are burned in the lower combustion section of the gasifier, and essentially all of the ash is converted to molten slag.

Steam and pulverized coal are injected into the upper (reducing) section of the gasifier where they encounter the hot gases leaving the combustion zone. Coal is devolatilized and is gasified by reaction with steam. The gas is cooled, mechanically cleaned and scrubbed to remove particulates prior to removal of H_2S by the Stretford gas/liquid contact process.

If the gasifier is air-blown, the product gas is expected to have a heating value of about 130 Btu/SCF. If it is oxygen blown, the heating value will be about 285 Btu/SCF.

TABLE 5.5.2. COMBUSTION ENGINEERING INC.

Facilty	Location	Owner(s) or Contractor	Status/Operating History
5 ton/hr process development unit	N.A.	Combustion Engineering Inc. (cosponsored by OCR and Consolidated Edison of New York)	Presently negotiating contract for construction of facility. Very little information available.

(Contact: John Anderson, Combustion Engineering Inc., Windsor, Conn. (203) 688-1911)

References:
(1) "Clean Energy from Coal - A National Priority", 1973 Annual Report (Calendar Year 1972), Office of Coal Research, Washington, D.C. (1973).
(2) "Coal Technology: Key to Clean Energy", Annual Report 1973-74, Office of Coal Research, Washington, D.C. (1974).

5.5.3 Foster-Wheeler (Foster-Wheeler Corporation)

Description of Process. Foster-Wheeler Corporation is designing an air-blown, entrained-flow gasifier for the production of clean, low-Btu gas as a fuel for conventional steam and combined-cycle power plants. A schematic is shown in Figure 5.5.3.

Dry, pulverized coal is fed into the second (upper) stage of the two-stage gasifier where it is entrained by hot gases from the first stage and devolatilzed at 1800-2100 F and 350 psig. The fixed-carbon in the coal is converted to a low density char which is separated from the gas stream by a series of cyclones and fed to the lower stage of the gasifier which is operated at slagging temperatures (2500-2800 F).

The nozzles through which char, air, and steam are fed into this stage are arranged so that slag is thrown out against the walls. The slag runs down the walls through a collector at the bottom and is quenched in a water bath.

It is expected that char will be essentially completely gasified in the lower stage.

The hot (1800 F) gas leaving the upper stage is cooled and then scrubbed to remove particulates and ammonia. Desulfurization is carried out in a closed-cycle wet-scrubbing system.

The clean, low-Btu gas is expected to have a heating value of 165 Btu/SCF.

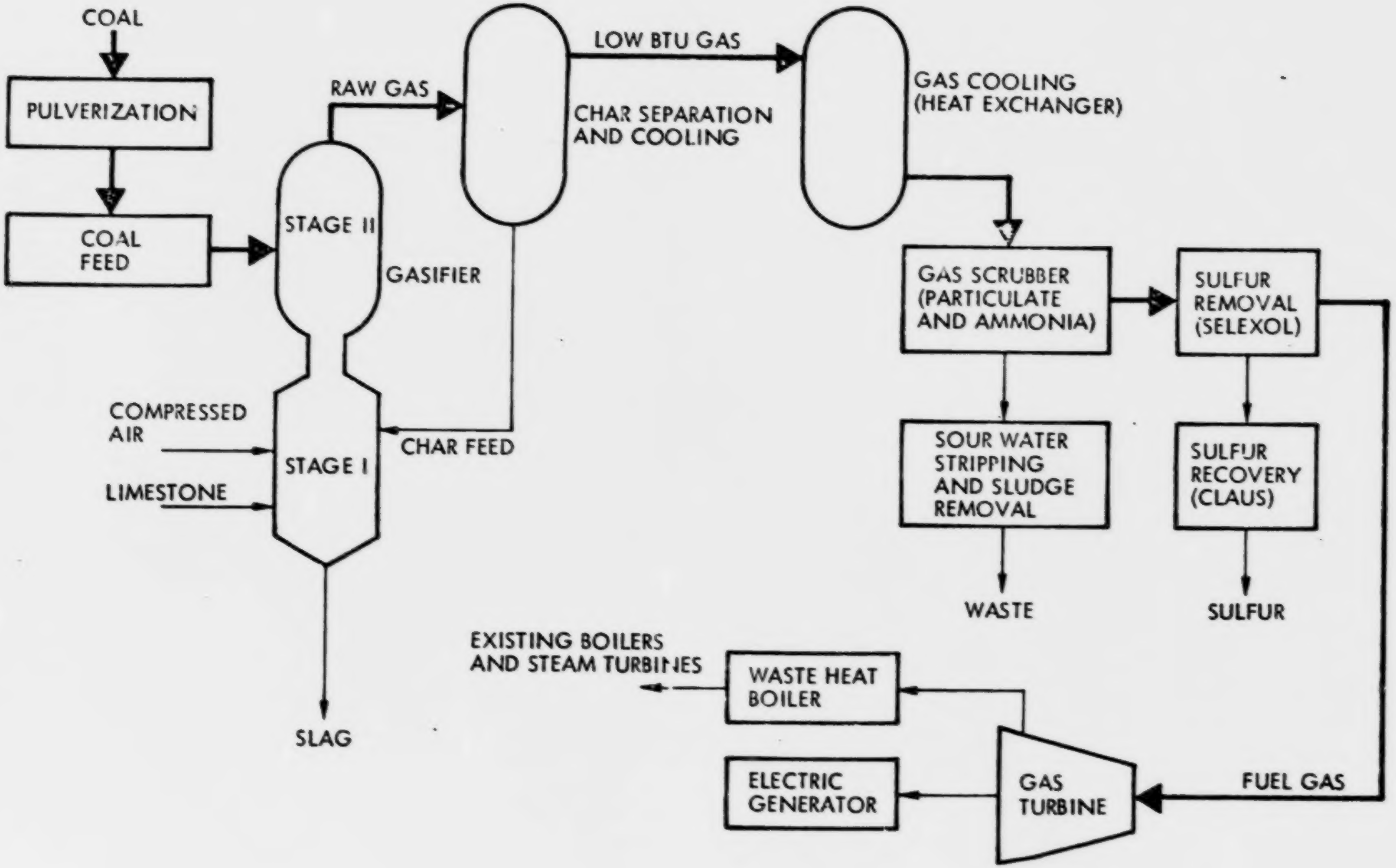


FIGURE 5.5.3. FOSTER WHEELER(3)

TABLE 5.5.3. FOSTER-WHEELER

Facilty	Location	Owner(s) or Contractor	Status/Operating History
5 ton/hr Pilot	South Dakota	Foster Wheeler (cosponsored and cofunded by OCR, Pittsburgh & Midway Coal Mining Co., Turbodyne and Northern States Power Co.)	Process design is completed. Construction is expected to begin in last quarter of 1974 and should be in operation by early 1977. Based on bench-scale studies with a 100 lb/hr unit. In phase I, low-Btu gas will go to modified existing boilers to generate power. In phase II, estimated to begin in early 1978, the gas will fuel a combined-cycle plant to generate about 130 MW of power.

(Contact: E. Damon, Foster-Wheeler Corp., Livingston, New Jersey (201) 533-3653)

References:
(1) Clean Energy from Coal: A National Priority, 1973 Annual Report, Office of Coal Research, pp 40-41, Washington, D.C. (1973).
(2) "Evaluation of Coal-Gasification Technology, Part II, Low- and Intermediate-Btu Fuel Gases", National Research Council, National Academy of Engineers, Washington D.C. (1973). (Process is listed under Pittsburgh and Midway Coal Company - the former project manager.)
(3) McCallister, R. A., and Ashley, G. C., "Coal Gasification to Produce a Low-Btu Fuel for a Combined-Cycle Power Station", Presented at American Power Conference, Chicago (April 29, 1974).

5.5.4 Garrett Flash Pyrolysis (Garrett Research and Development CO.)

Description of Process. Pulverized coal is fed to the gasifier which operates at about 50 psi (Figure 5.5.4). The coal is rapidly heated to 1500 to 1700 F by hot recycle char in entrained flow. Because of the short residence time in the pyrolysis zone (two seconds or less), gas-phase cracking reactions are minimized with a resultant high direct yield of hydrocarbons. At 1600 F, typical gas composition is reported to be as follows: H_2, 35.3; CO, 22.4; CO_2, 9.1; CH_4, 18.8; C_{2+} (Predominantly ethene), 14.1 (Vol. %; nitrogen-free basis).

Char is separated from the gas leaving the pyrolysis reactor by a series of cyclones and sent to the char heater. A portion of the char is partially burned with air, and after separation from combustion gas, is recirculated to the pyrolysis reactor to provide process heat. This latter feature makes it possible to produce nitrogen-free gas of about 600 to 650 Btu/SCF without using pure oxygen.

In addition to the product gas, the Garrett process also produces high yields of char and tar. (At 1600 F the relative yields (by weight) of gas, char, and tar are about 30, 56, and 14, respectively (1).)

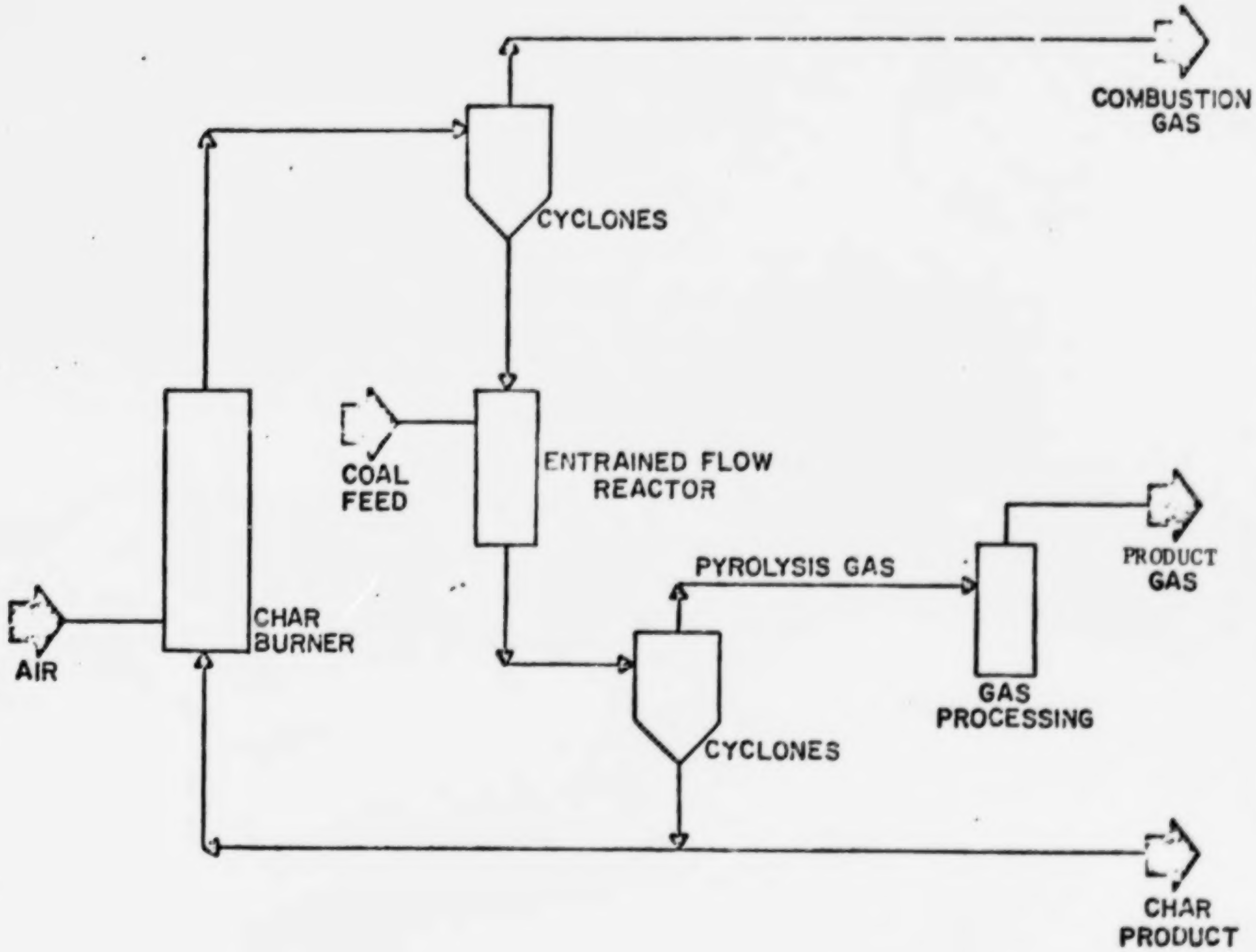


FIGURE 5.5.4. GARRETT FLASH PYROLYSIS PROCESS(4)

TABLE 5.5.4. GARRET FLASH PYROLYSIS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
10 ton/hr Pilot	LaVerne, Ca.	Garret Research and Development (research subsidiary of Occidental Petroleum)	Proposed. Seeking support. A bench scale pyrolysis unit (50 lb/hr) has been in operation since Jan. 1973 (2).

(Contact: D. E. Adams, Garrett Research and Development, La Verne, Ca., (714) 593-7421)

References:

(1) McMath, H. G., Lumkin, R. E., and Sass, A., "Production of Gas from Western Sub-Bituminous Coals by the Garret Flash Process", Presented at the 66th Annual Meeting AICHE Philadelphia, Pa. (November 1973).

(2) McMath, H. G., Lumkin, R. E., Longanbach, J. R., and Sass, A., "A Pyrolysis Reactor for Coal Gasification", Chemical Engineering Progress, Vol. 70, No. 6, pp 72-3 (June 1974).

(3) Adams, D. E., Sack S., and Sass, A., "Coal Gasification by Pyrolysis", ibid pp 74-75.

(4) Adams, D. E., Sack S., and Sass, A., "The Garret Pyrolysis Process", presented at the 66th Annual Meeting AICHE, Philadelphia, Pa. (November 15, 1973).

5.6 Entrainment/Slagging Reactors

Operating temperatures are high enough to produce slagging conditions. A large fraction of the ash (more than 50 percent in the Koppers-Totzek process) flows down the gasifier walls as molten slag into a slag quench tank; the remainder of the ash leaves the gasifier as fine fly ash entrained in the exit gas. The higher temperatures lead to higher specific gasification rates.

5.6.1 Koppers-Totzek (Koppers Engineering and Construction)

Description of Process. The Koppers-Totzek gasifier, shown schematically in Figure 5.6.1, is an oxygen-blown, entrained-flow system which has been used extensively for production of hydrogen in the synthesis of ammonia.

Dried, pulverized coal is screw-fed into pairs of opposing burners arranged so that their jet discharges converge. Gasifiers may employ either two or four such burner heads.

Reaction temperatures at the burner discharge are in the range 3300 to 3500 F, while the exit gas temperature is at about 2750 F. Under these conditions only gaseous products are generated (no tars, condensable hydrocarbons,or phenols).

Approximately half of the coal ash drops out as slag into a slag quench tank below the gasifier while the other half leaves as fly ash with the product gas. When necessary, molten ash particles can be solidified by water sprays to prevent buildup in tubes of the waste-heat boiler. After further cooling, the gas is purified by conventional methods.

A major advantage of this process is the wide variety of acceptable feedstocks (all ranks of coal, char, petroleum coke, tars, heavy residuals, light to heavy oils and pumpable slurries of carbonaceous materials in hydrocarbon liquids).

The present generation gasifiers operate at atmospheric pressure but units capable of operating at pressures up to about 15 atms are presently under development.

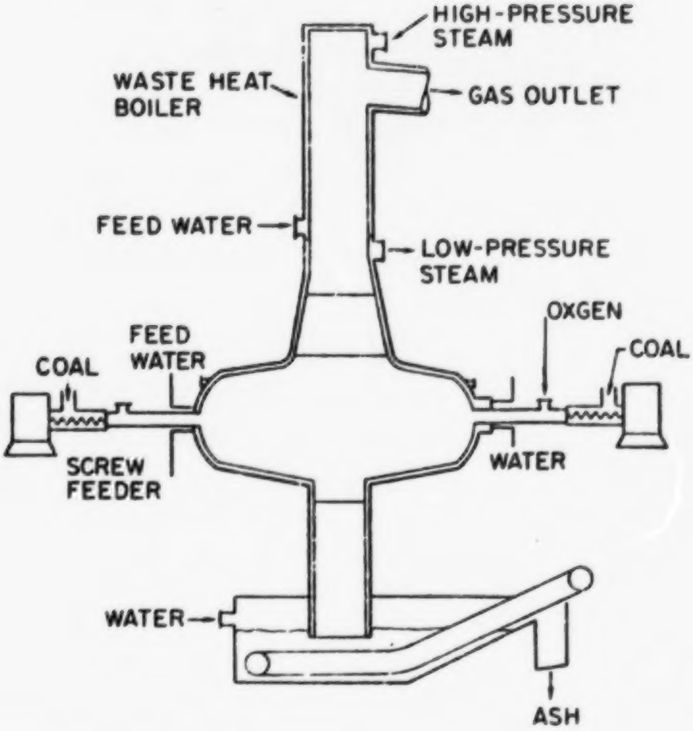


FIGURE 5.6.1. KOPPERS-TOTZEK GASIFIER(3)

TABLE 5.6.1. KOPPERS-TOTZEK: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
10.4 x 10^6 SCF/day	Oulu, Finland	Typpi Oy	5 gasifiers (3 ordered in 1950; 2 ordered in 1955) Produce synthesis gas from coal dust, oil and peat.
7.8 x 10^6 SCF/day	Tokyo, Japan	Nihon Suiso Kogyo Kaisha, Ltd.	3 gasifiers produce synthesis gas from coal dust (ordered in 1954)
15.5 x 10^6 SCF/day	Puentes de Garcia Rodriguez, Spain	Empresa Nacional "Calvo Sotelo" de Combustibles Liquidos y Lubricantes	4 gasifiers (3 ordered in 1954; 1 ordered in 1961) produce synthesis gas from lignite dust and/or Naptha.
6.55 x 10^6 SCF/day	Zandvoorde, Belgium	S.A. Union Chimique Belge	2 gasifiers produce synthesis gas from bunker-C-oil, convertible to coal dust (ordered in 1955).
6.3 x 10^6 SCF/day	Estarreja, Portugal	Amoniaco Portugues S.A.R.L.	2 gasifiers produce synthesis gas from heavy gasoline but convertible to coal dust (ordered in 1956).
38.6 x 10^6 SCF/day	Ptolemais, Greece	Government of Greece	6 gasifiers (4 in 1959; 1 in 1969; 1 in 1970) produce synthesis gas from lignite dust and bunker-C oil.
8.07 x 10^6 SCF/day	Lampang, Thailand	Chemical Fertilizer Co., Ltd.	1 gasifier produces synthesis gas from lignite dust (ordered in 1963).
28.85 x 10^6 SCF/day	Kutahya, Turkey	Azot Sanayii T.A.S.	4 gasifiers produce synthesis gas from lignite dust (ordered in 1966).
13.4 x 10^6 SCF/day	East Germany	VEB Germania, Chemieanlagen und Apparatebau	2 gasifiers produce gas for hydrogenation from vacuum residue and/or fuel oil (ordered in 1966).
7.98 x 10^6 SCF/day	Zambia, Africa	Industrial Development Corp., Zambia	1 gasifier produces synthesis gas from coal dust (ordered in 1967).
74.45 x 10^6 SCF/day	Ramagundam, India	Fertilizer Corp. of	3 gasifiers produce synthesis gas from coal dust (ordered in 1969).
"	Talcher, India	"	As above (ordered in 1970).
"	Korba, India	"	As above (ordered in 1972).
80 x 10^6 SCF/day	Modderfontein, South Africa	AE & CI Ltd.	6 gasifiers produce synthesis gas from coaldust (ordered in 1972).

(Contact: J. F. Farnsworth, Koppers Engineering and Construction, Pittsburgh, Pa., (412) 391-3300)

TABLE 5.6.1. (Continued)

References:

(1) "Evaluation of Coal-Gasification Technology, Part II, Low- and Intermediate-Btu Fuel Gases", National Research Council and National Academy of Engineering, Washington, D.C. (1973).

(2) Farnsworth, J. F., Leonard, H. F., Mitsak, D. M., and Wintrell, R., "Production of Gas from Coal by the Koppers-Totzek Process", Symposium on Clean Fuels from Coal, pp 143-162, Institute for Gas Technology, Chicago (1973).

(3) Indian Government Sponsored Study of Commercial Coal Gasification Processes, V. N. Kasturiranzan, M. Satyapal, R. R. Iyer, D. G. Rao and S. B. Chatterji, reproduced by Koppers Engineering and Construction Company, Pittsburgh (1973).

(4) Magee, E. M., Jahnig, C. E., and Shaw, H., "Evaluation of Pollution Control in Fossil Fuel Conversion Processes, Section 1: Koppers-Totzek", Prepared for Office of Research and Development, USEPA, Washington, D.C. (January 1974).

5.6.2. Texaco

Description of Process. A simplified schematic of the Texaco gasifier is shown in Figure 5.6.2. Coal is fed to the gasifier as a slurry. The coal-water slurry is pumped through a preheater in which the water is vaporized and the mixture heated to about 1000 F. The steam-coal mixture is then blown into the top of the gasifier, and preheated oxygen is fed through a separate water-cooled nozzle. The steam-coal ratio is controlled by a cyclone separator ahead of the gasifier. Temperatures in the reaction zone range from 2000-2500 F and the operating pressure is 400 psi.

Slag is withdrawn from the bottom of the unit and quenched with water. Heat from the gas leaving the reactor is recovered by a waste heat boiler.

The gasifier can be blown with either oxygen or air.

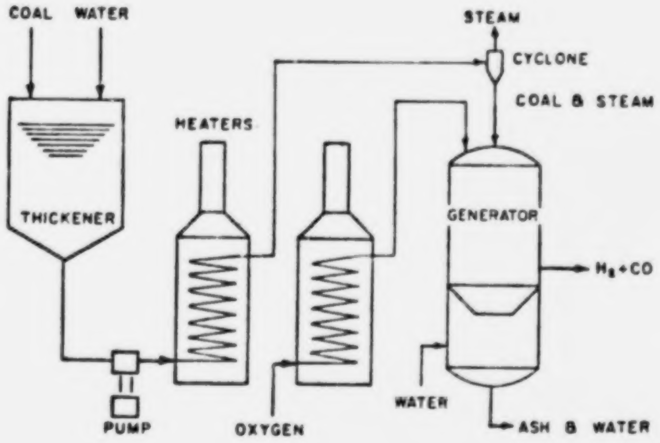


FIGURE 5.6.2. SIMPLIFIED FLOW DIAGRAM OF THE TEXACO(1) GASIFIER

TABLE 5.6.2. TEXACO: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
			At the present time, Texaco has no commercial units in operation that use coal as a feedstock. However, Texaco has previous pilot experience in coal gasification operations (Morgantown, W.Va.) and a semi-commercial unit was in operation for a number of years that produced synthesis gas from coal.
	(Contact: Peter L. Paul, President, Texaco Development Corp., New York, N.Y. (212) 953-6734		

References:

(1) Von Fredersdorff, C. G., Elliot, M. A., "Coal Gasification" in Chemistry of Coal Utilization, H. H. Lowery, Editor, Supplementary Volume, John Wiley and Sons, Inc., N.Y., pp 982-3 (1963).

(2) Bituminous Coal Research, Inc., "Gas Generator Research and Development Survey and Evaluation - Phase One", R&D Report No. 1., U.S. Dept. of Interior, O.C.R., Washington (1965).

5.6.3 Babcock & Wilcox

Description of the Process. A schematic of the commercial-scale unit, which was installed at the Belle, West Virginia plant of the du Pont Company, is shown in Figure 5.6.3. The refractory-lined gasifier was cylindrical with a primary and a secondary reaction zone. Pulverized coal was swept into burners in the primary zone with steam and oxygen. The lower or primary zone operated at slagging temperatures and molten slag was continuously tapped from the bottom of the gasifier. The secondary zone operated at lower temperatures. A waste-heat boiler recovered heat from the hot (about 2000 F) exit gas.

This process accepts all types of coal. It produces a gas which is primarily carbon monoxide and hydrogen (in nearly equal yields). The methane yield is less than one percent.

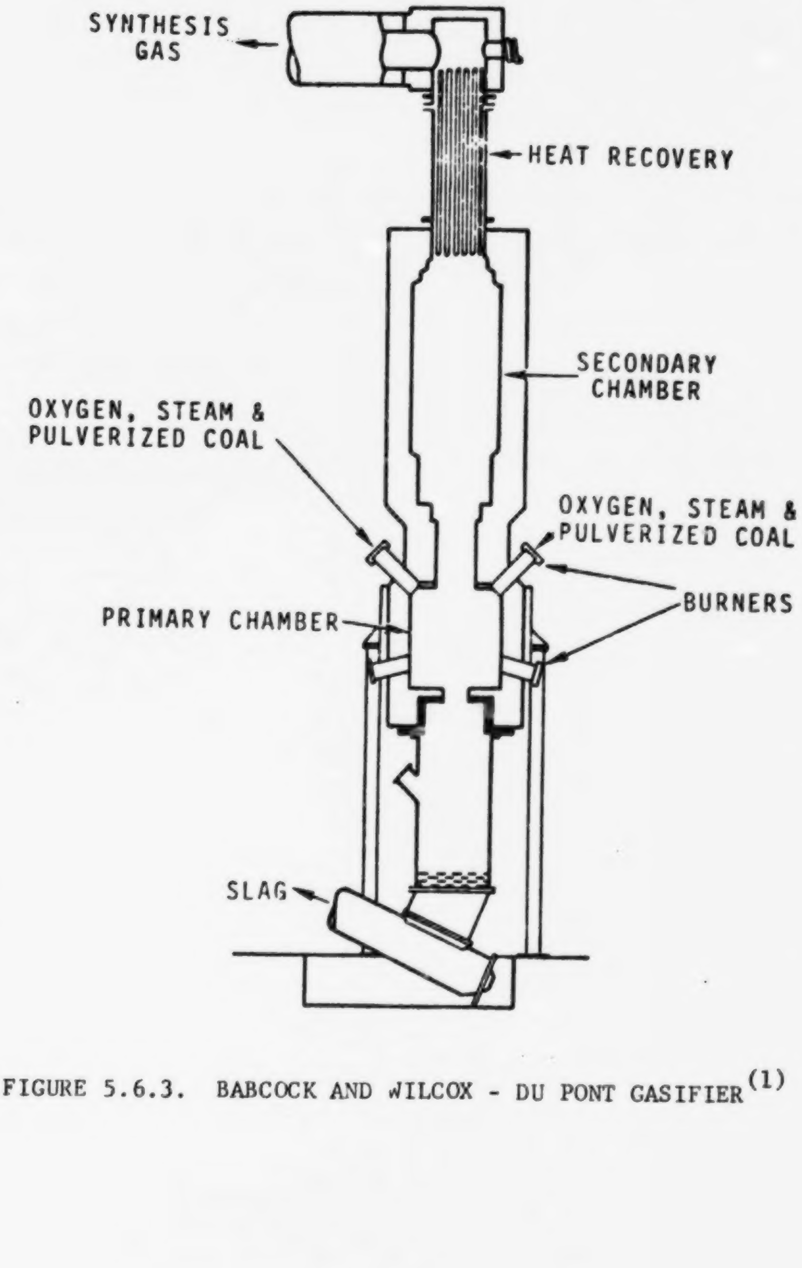


FIGURE 5.6.3. BABCOCK AND WILCOX - DU PONT GASIFIER(1)

TABLE 5.6.3. BABCOCK & WILCOX: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
500 lb/hr process development unit	Morgantown, W.Va.	Designed and constructed by B&W at U.S. Bureau of Mines in Morgantown	Began operating in Summer of 1951. Operational data have been published.(2)
3,000 lb/hr Pilot	Belle, W.Va.	Installed by duPont	Began operation in fall of 1951. Provided performance data for design of commercial unit. Operational data have been published.(2)
17 TPH Commercial Plant for Production of Synthesis Gas	Belle, W.Va.	"	Plant was operated for 1 year. Operational data has not been published. Plant has been dismantled.

(Contact: Sidney Katell, U.S. Bureau of Mines, Morgantown, W.Va (304) 599-7000)

References:

(1) VonFredersdorff, S. C. and Elliot, M. A., "Coal Gasification", in Chemistry of Coal Utilization, H.H. Lowry (Editor), Supplementary Volume, John Wiley and Sons, Inc., New York, p 974 (1963).

(2) Grossman, P. R. and Curtis, R. W., "Pulverized-Coal-Fired Gasifier for Production of Carbon Monoxide and Hydrogen", ASME Transactions, 76, 689-95 (1954).

5.7 Molten Bath

Gasification of coal has also been carried out in baths of molten iron and molten salts. An important consideration common to such systems is the ability to accept all types of coal. Also, a significant fraction of the sulfur may be removed in the bath. Unfortunately, such processes usually involve formidable materials problems.

5.7.1 Molten Iron (Applied Technology Corp.)

Description of Process. There are three variations of the basic process: The Two-stage Coal Combustion Process which produces a 190 Btu/SCF fuel gas, the PATGAS Process which produces a 315 Btu/SCF gas, and the ATGAS Process which produces SNG (the Atgas Process is described in Section 6.10).

In the Two-stage process coal and limestone are injected with compressed air into a molten iron bath at 2500 F (Figure 5.7.1). Fixed carbon and sulfur dissolve and are retained by the iron, while the coal volatiles crack and appear as offgas. Dissolved carbon is gasified by reaction with additional air which is injected slightly below the surface of the molten iron to yield carbon monoxide.

To prevent build-up of sulfur in the molten iron, dissolved sulfur is continuously transferred to a molten, lime-bearing slag floating on the surface of the iron. The slag, containing 4 to 8 percent sulfur as calcium sulfide, and all of the coal ash, is continously removed and desulfurized. The raw gas (30 percent CO, 15 percent H_2, 55 percent N_2) can be used as a low-Btu (190 Btu/SCF) fuel gas.

In the PATGAS Process steam is used as the carrier for coal and limestone,and oxygen (rather than air) is injected into the molten bath. In this case the offgas is 63.5 percent CO, 36 percent H_2 and 0.5 percent N_2, and has a heating value of 315 Btu/SCF.

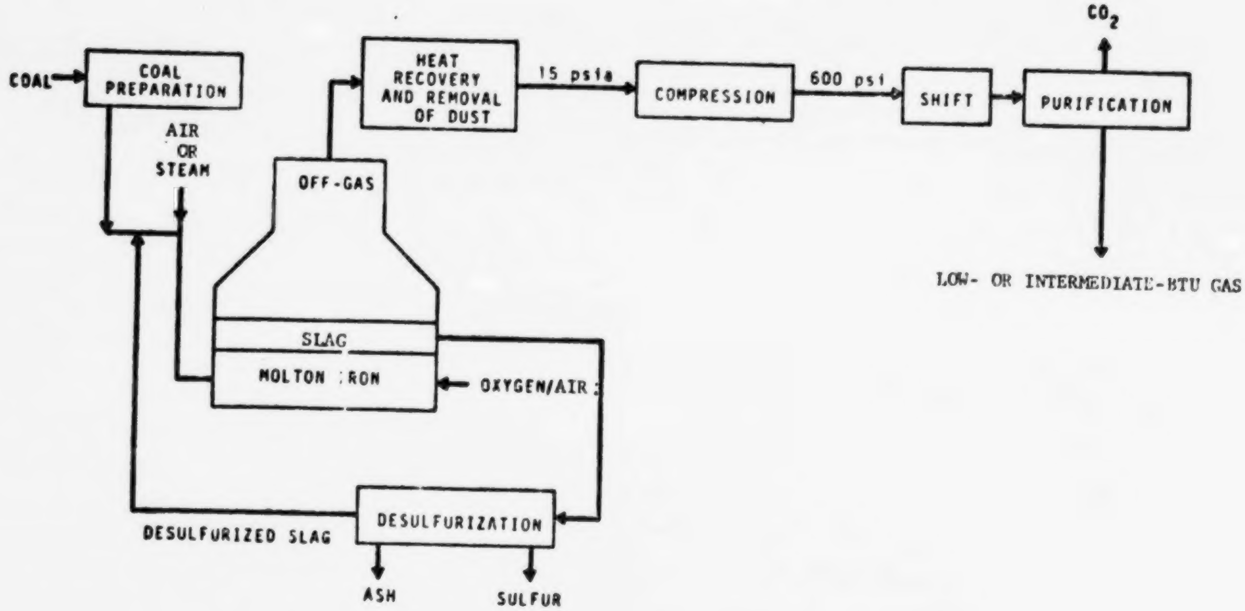


FIGURE 5.7.1. MOLTEN IRON PROCESS(1)

TABLE 5.7.1. MOLTEN IRON: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
-------	-------	Applied Technology Corp.	Pre-pilot stage. Studies have involved a 25 inch I.D. induction furnace (4000 lb capacity) to simulate the gasifier, and use of air to produce low-Btu fuel gas. The off gas handling system was equipped to permit continuous analysis for SO_2, NO_2, NO, H_2, O_2, CO_2 and CO. Expendable, non-cooled ceramic lances were used for injection. Results indicate a boiler feed gas with less than 50 ppm SO_2 can be generated from high-S coal (3.5 percent)$^{(1)}$

(Contact: Ronald J. McGarvey, Applied Technology Corp., Pittsburgh, Pa (412) 782-0682)

References:

(1) LaRosa, Paul and McGarvey, Ronald J., "Fuel Gas from Molten Iron Coal Gasification", Institute of Gas Technology Symposium on Clean Fuels, pp 285-3000, Chicago (1973).

(2) "Evaluation of Coal Gasification Technology, Part II, Low-and Intermediate-Btu Fuel Gases, National Research Council, National Academy of Engineers, Washington, D.C. (1973).

(3) "Clean Energy from Coal: A National Priority", 1973 Annual Report (for Calendar Year 1972) Office of Coal Research, Washington, D.C. (1973).

5.7.2 Kellogg Molten Salt (M. W. Kellogg Company)

Description of Process. When intermediate Btu-gas is to be produced, crushed, dried coal and sodium carbonate from lock hoppers are carried into the molten-salt gasifier by preheated oxygen and steam. (Figure 5.7.2). The gasifier operates at 1700 F and about 1200 psi; however, lower pressure can be used if high yields of methane are not required.

Under these conditions the major gasification reactions are coal with steam, which is catalyzed by sodium carbonate, partial-combustion of coal and non-catalytic methanation.

A bleed stream of molten salt containing ash is withdrawn from the bottom of the gasifier and quenched with water. Sodium carbonate is dissolved and the ash is separated by filtration. The filtrate is carbonated with carbon dioxide from the purification system to precipitate sodium bicarbonate. The bicarbonate is filtered off and heated to regenerate the carbonate salt for recycle to the gasifier.

After the raw gas is processed to recover heat and entrained salt, carbon dioxide and the remaining sulfur are removed by the Selexol process. (A significant fraction of the sulfur is removed by reaction with molten carbonate in the gasifier.)

Gasification can also be carried out with steam and air to produce a low-Btu fuel gas. In this case, the gasifier can be operated at much lower pressures.

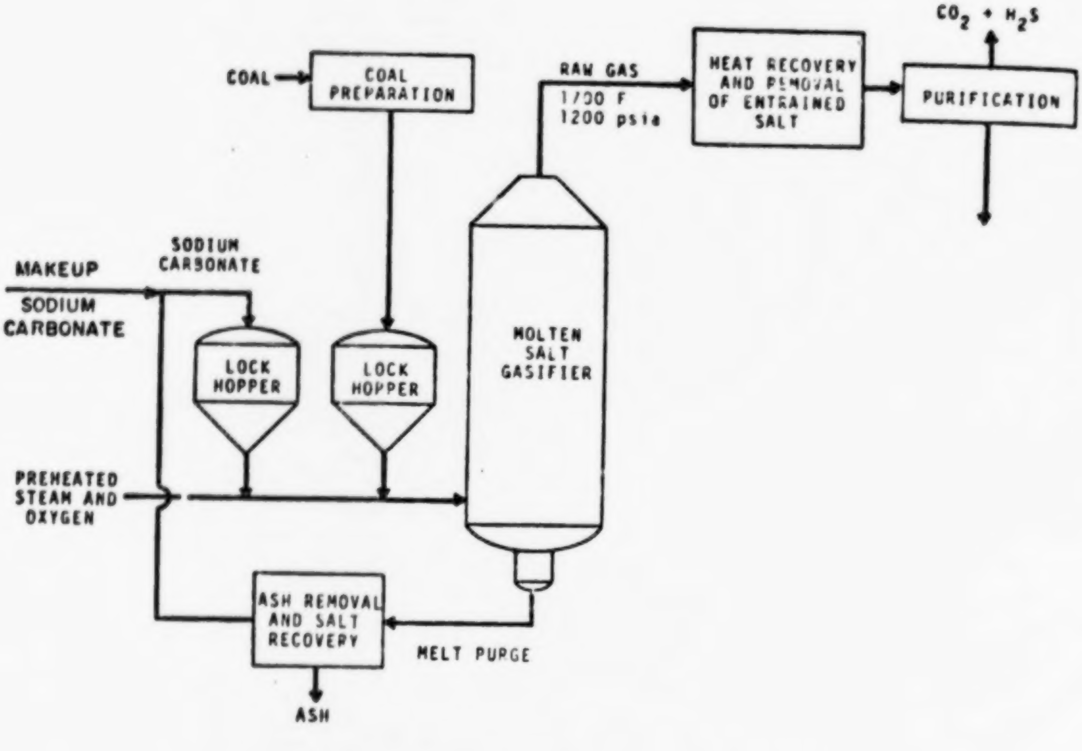


FIGURE 5.7.2. KELLOGG MOLTEN-SALT PROCESS(2)

TABLE 5.7.2. KELLOGG MOLTEN SALT: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
-------	------	M.W. Kellogg Company	Bench scale studies only. The molten salt creates serious corrosion problem, but these have been solved by use of an aluminum oxide refractory, Monofrax A. Report no corrosion after 600-700 hrs continuous operation. A process development unit is planned. Preliminary flow sheets and cost estimates have been made. Seeking support. Gasification of coal in molten K_2CO_3 to produce a fuel suitable for magnetohydrodynamic systems is also being studied by Kellogg with support from OCR (4).
(Contact: A. E. Cover, M. W. Kellogg Company, Houston, Texas, (713) 626-5600).			

References:

(1) Cover, A. E. and Schreiner, W. C., "The Kellog Molten Salt Process", Presented at the 4th Conference on Synthetic Fuels from Coal, Oklahoma State University, Stillwater, Oklahoma (1974).

(2) Cover, A. E., Schreiner, W. C., and Skaperdas, G. T., "The Kellogg Coal Gasification Process: Single Vessel Operation", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 273-284, Chicago (1973).

(3) Evaluation of Coal-Gasification Technology, Part II, Low- and Intermediate-Btu Fuel Gases, National Research Council, National Academy of Sciences, Washington, D.C. (1973).

(4) "Coal Technology: Key to Clean Energy", Annual Report 1973-74, Office of Coal Research, p 62, Washington, D.C. (1974).

5.7.3 Atomics International Molten Salt

Description of Process. This process involves gasification of coal with air in molten sodium carbonate to produce a clean low-Btu fuel gas.

Crushed coal and compressed air together with a small amount of sodium carbonate make-up are fed into the molten salt bath which operates at 1800 F and a pressure of 5 to 10 atmospheres. Coal is partially oxidized by air to CO, CO_2 and H_2O with complete release of coal volatiles into the product gas stream. These reactions take place rapidly at the relatively low operating temperature primarily because of the catalytic effect of sulfates and sulfides dissolved in the melt.

Because sulfur and ash from the coal are retained in the melt, the off-gas can be burned in a conventional gas-fired boiler without costly clean-up processes.

The sulfur and ash content of the melt will be optimized to provide satisfactory operation of the bath while minimizing the flow rate of the sidestream of melt which is taken to the regeneration system for removal of sulfur and ash, and regeneration of sodium carbonate. Regeneration involves quenching with enough water to dissolve the salts, filtering off the ash, and carbonation with product gas to regenerate sodium carbonate and release hydrogen sulfide. Sodium carbonate is crystallized from the regenerated solution and dried; hydrogen sulfide is converted to elemental sulfur by the Claus process.

The product gas has a heating value of about 150 Btu/SCF.

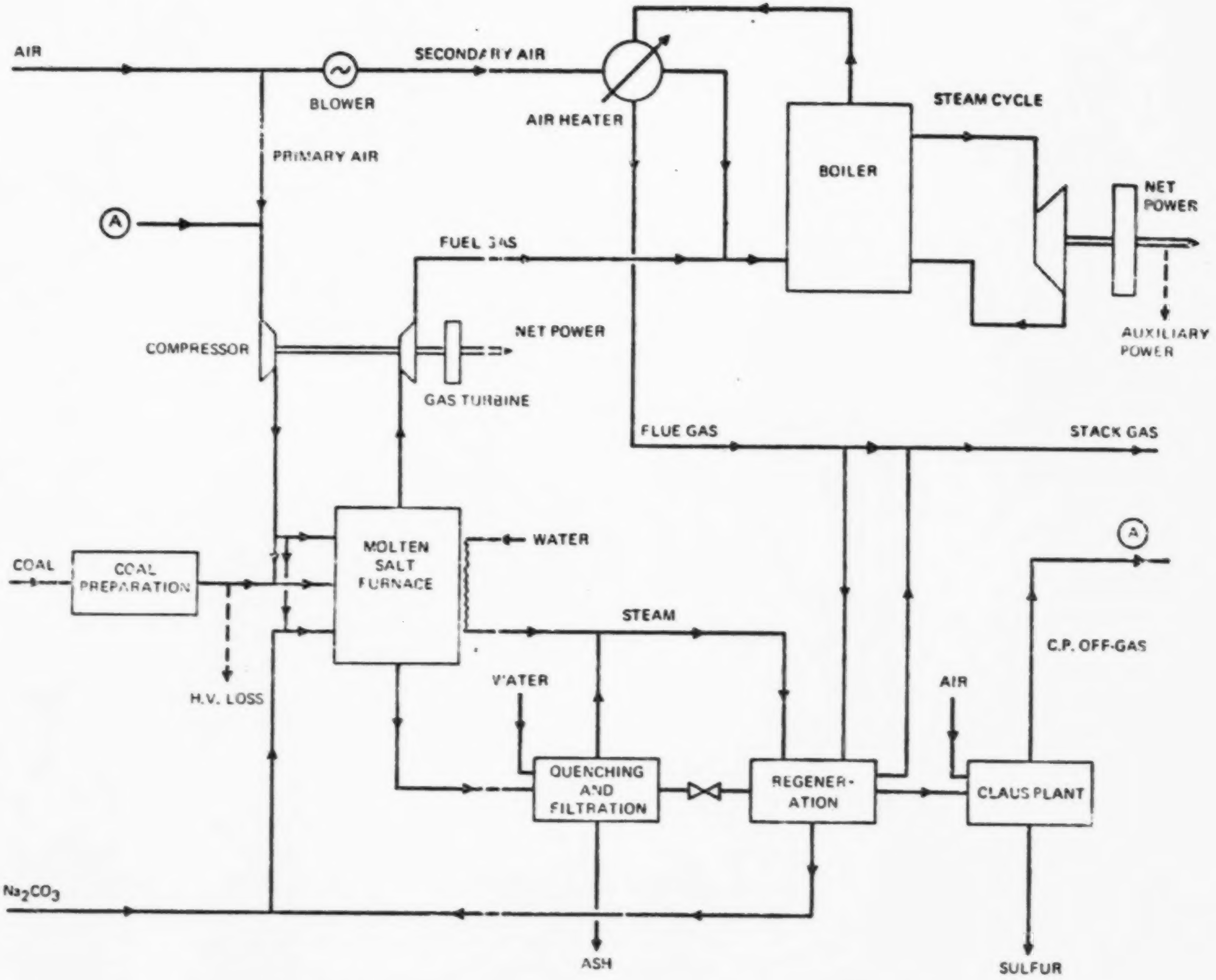


FIGURE 5.7.3. ATOMICS INTERNATIONAL MOLTEN SALT GASIFICATION PROCESS(1)

TABLE 5.7.3 ATOMIC INTERNATION MOLTEN SALT: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
5 Ton/hr Pilot	Norwalk Station, Conn.	Atomics International	Conceptual design. Seeking Funding. Bench scale tests on 3 ft I.D. gasifier. Batch process. Ash build-up determines length of run. Completed runs at atmospheric pressure with processing of 170 lb/hr; next runs will involve 500 lb/hr at 5 atm.

(Contact: Charles A. Trilling, Atomics International, Canoga Park, Ca., (213) 341-1000)

References:
(1) Trilling, Charles A., "Molten Salt Process for the Gasification of Coal", Prepared for presentation at the Workshop on Materials Problems and Research Opportunities in Coal Conversion, Ohio State University (April 16-18, 1974).
(2) "Evaluation of Coal Gasification Technology, Part II, Low- and Intermediate-Btu Fuel Gases", National Research Council, National Academy of Engineering, Washington, D.C. (1973).

5.8 Underground Gasification

Description of Process. Underground gasification of coal is the burning of coal *in situ* in the presence of air, oxygen, steam-air, or steam-oxygen mixtures introduced into the seam by boreholes or shafts. The process permits recovery of a low-Btu gas without recourse to mining operations. The chemistry is essentially the same as that involved in the other processes described in this chapter.

The technology of underground gasification has been thoroughly reviewed in a recent report to the U.S. Bureau of Mines prepared by A. D. Little, Inc. (44).

The English chemist Sir William Ramsey carried out small-scale studies of underground gasification prior to World War I, and the Russians did rather extensive work in the period from 1933 to about 1965. Major research efforts were also concentrated in the United Kingdom and in the United States during the period following World War II. However, research activity in the area of underground gasification was essentially abandoned in all three countries by the early sixties.

The studies did demonstrate the technical feasibility of underground gasification-at least on a relatively small scale, but the gas produced was generally of poor quality.

More recently, the U.S. Bureau of Mines has shown renewed interest in underground gasification, and experimental studies are now being conducted at a site near Hanna, Wyoming which will explore the technological, economic,and environmental feasibility of the underground gasification of western subbituminous coal (46).

TABLE 5.8 UNDERGROUND GASIFICATION

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Commercial	USSR	Government	By July 1941, when USSR entered WW II gas was being produced from 5 underground gasification installations. Further development was interrupted by the war. By 1958, USSR was producing 27 x 10^{12} SCF low-Btu gas; however, interest in gasification declined in the early sixties apparently because of increased availability of natural gas (2).
Experimental	Gorgas, Alabama	US Bureau of Mines and Alabama Power Co.	First of a long series of experiments on in situ gasification were begun in early 1947. Studies were made with air, O_2-enriched air with and without steam and with O_2-steam mixtures. Later (1951) studied methods of increasing seam permeability (electrolinking and hydraulic fracturing). Work at Gorgas was terminated in 1959. Present US activity is centered at Hanna, Wyo. (see later entry). (2)
Experimental	Banco-Casino, Italy	Socogaz and Mineraria, Valdarno	Underground gasification tests in lignite seam (15-60 ft thickness). Early tests (1947) produced 7 MMSCF/D of gas with average heating value of 100 Btu/SCF. No longer in operation.(2)
Experimental	Wandre, Belgium	Soxogaz, Charbonnages de France and Charbonnage de Pologne (Poland)	Experimental work on semi-anthracite seam about 3 ft thick was carried out in 1948-50. Results were poor and work was discontinued. (50 percent of the Belgian coal reserves is in seams too thin for conventional mining).(2)
Experimental	Newman Spinney and Bayton (England)	National Coal Board	Work began in 1949 and continued through 1960. Culminated with a pilot electrical-generating station operating on low-Btu gas from the Newman Spinney installation. The plant generated a total of about 3.5 KWH from 238 MMSCF of gas with an average heating value of 58.8 Btu/SCF (fluctuated between 40 and 84 Btu/SCF).(2)
Experimental	Hanna, Wyo.	US Bureau of Mines	Ignition for 1st trial was in March 1973, and continued through April 1974, 30 ft seam. Forward burning did not go well. Switched to backward burning in May with considerably greater success. Between Sept. and Feb. gas was produced at a rather steady rate of about 1.6 MMSCFD (dry). The heating value was about 130 Btu/SCF. There were no significant process problems. Preliminary estimates indicate about 65 percent coal recovery (to be verified by seismic and coring studies).

TABLE 5.8 (Continued)

Facilty	Location	Owner(s) or Contractor	Status/Operating History
			First phase involved air injection. Second phase, which will run about 3-4 months, will involve injection of O_2-enriched air and pneumatic linking. If results are as successful as Phase I, it will have been demonstrated that it is possible to sustain a reasonable steady flow of gas of approximately uniform composition.(4)

References:
(1) Elder, J. L., "The Underground Gasification of Coal", Chemistry of Coal Utilization, H. H. Lowry, Editor, Supplementary Volume, John Wiley and Sons, Inc., New York, pp 1023-1040 (1963).
(2) A. D. Little, Inc., "A Current Appraisal of Underground Coal Gasification", U.S. Dept. of the Interior, Bureau of Mines, Washington, D.C. PB 209 274, NTIS, Springfield, Va., 280 p (April 1972).
(3) Nadkarni, R. M., Bliss, Charles and Watson, W. I., "Underground Gasification of Coal", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 611-637, Chicago (1973).
(4) Private Communication.
(5) Schrider, Leo A., and Pasini, J., Underground Gasification of Coal--Pilot Test, Hanna, Wyoming, presented at the AGA Fifth Synthetic Pipeline Gas Symposium (October 1973).

5.9 Gasification of Refinery Residues

Although this section will be devoted to processes developed primarily or exclusively for the gasification of low-cost refinery residues or "bottoms", it should be noted that many of the processes described earlier in this chapter can - in some cases without significant modification - be used to gasify residual oils.

Commercial scale Koppers-Totzek gasifiers in Belgium and Greece have operated with Bunker-C oil feed to produce synthesis gas for a number of years (Table 5.6.1). All of the molten bath processes can accept high sulfur residual oils, and Covers and Schreiner(*) briefly discussed application of the Kellogg Molten Salt Process to the cracking of heavy oils to produce liquid and/or gaseous products. Product distribution for a given feed depends upon bath temperature,which can be controlled by addition of lithium and potassium carbonates to the sodium carbonate to lower the freezing point.

Also, it should be added that the technology for production of fuel gases, in particular SNG from naptha and kerosine fractions boiling up to 600 F, is well developed. A complete list of plants for production of SNG from petroleum, announced as of April 15, 1973, has been published.(47)

(*) Cover, A.E. and Schreiner, W. C., The Kellogg Molten Salt Process, Presented at the 4th Conference on Synthetic Fuels from Coal, Oklahoma State University, Stillwater, Oklahoma (May 1974).

5.9.1 Flexicoking (Exxon Corporation)

Description of Process. A simplified schematic of the Flexicoker is shown in Figure 5.9.1. Residuum feed injected into the reactor is in part thermally cracked to a wide range of volatiles, and in part converted to coke which deposits on fluidized circulating coke particles. Heat is supplied to the reactor by recirculating a stream of coke particles from a heater vessel. Volatiles leaving the reactor are quenched in the scrubber section. Materials boiling above about 950 F condense and can be recycled to the coking reactor.

Coke produced in the reactor flows to the heater and is partially devolatilized at 1150 F. A coke-stream also goes from the heater to the gasifier where it is reacted at an elevated temperature with air, or oxygen and steam to produce the product gas (coke gas).

The coke gas from the gasifier and the light hydrocarbons produced by devolatilization in the heater are cooled and coke fines are removed for further processing. The clean product gas has a heating value of about 100 to 130 Btu/SCF if air is used for gasification.

The process is expected to convert 98 percent by weight of a vacuum residuum to gaseous and liquid products. Greater than 99 percent of the metals in the feed are concentrated in a 2 percent solids purge. Approximately 95 percent of the total sulfur in the residuum can be recovered as elemental sulfur by commercially available processes.

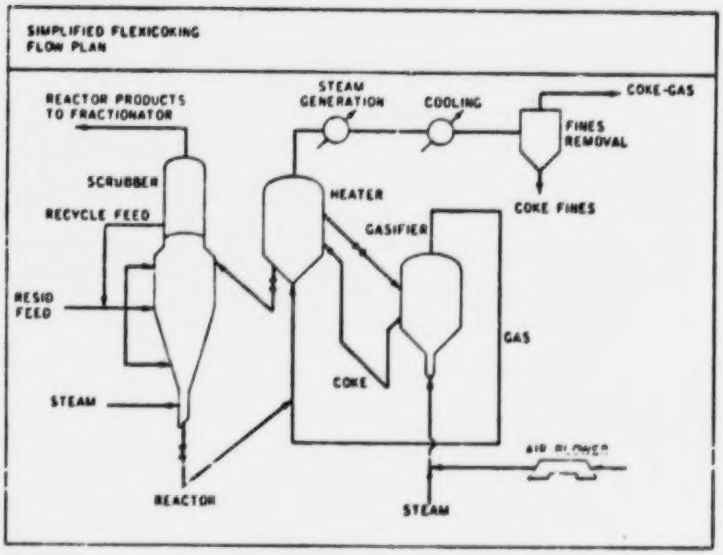


FIGURE 5.9.1. FLEXICOKING (Exxon Corporation)(1)

TABLE 5.9.1. FLEXICOKING: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
2 bbl/day Process Development Unit	Baton Rouge, La.	Exxon Corp.	This integrated coking/gasification unit has been commissioned and is in operation. Data indicate that typical coke desulfurization levels are 70 percent or higher.
750 bbl/day Prototype Unit	Baytown, Texas	"	Starting up.
21 x 10^3 bbl/day Commercial Unit	Kawasaki, Japan	TOA Oil of Japan	Under construction. Due onstream in 1975.
Commercial Unit	Nagoya, Japan	"	Being designed.

(Contact: M. P. Margolis, Exxon Research and Engineering, Florham Park, N.J. (201) 474-0100)

References:

(1) Matula, J. P., Weinberg, H. N. and Weissman, W., "Flexicoking: An Advanced Fluid Coking Process", American Petroleum Institute, Proceedings, Division of Refining, 37th Midyear Meeting, New York (May 8-11, 1972).

(2) Rionda, J. A., Jr., Bodnick, S., Kett, T. K., Metrailer, W. J., Savage, H. R., and Duir, J. H., "Recent Advances in Residua Processing", presented at National Petroleum Refiners Association Annual Meeting (April 2, 1974).

5.9.2. Texaco Partial Oxidation (Texaco Oil Company)

Descripiton of the Process. Oil and recycled soot undergo partial oxidation in a refractory-lined generator with either air or oxygen. Steam is added to moderate the temperature. An especially developed feed system for oil, steam and oxidant is carefully matched to a highly efficient burner which results in very effective mixing and combustion.

In addition to oxidation of hydrocarbons to hydrogen and carbon monoxide, steam cracking, hydrocracking and carbonization reactions also take place in the generator. The relative importance of these reactions is determined by such factors as operating temperature and pressure, flow rate, and ratios of oxidant to hydrocarbon and steam to hydrocarbon feed.

Commercial units have been operated at pressures up to 1200 psi and large-scale pilot units have been operated at pressures up to 2500 psi. Temperatures range from 1800 to 3000 F.

Generators can be operated in the direct water quench mode (Figure 5.9.2) or with a waste-heat boiler between the generator and scurbber. The direct water quench approach is usually favored if processing of the gas involves shift conversion.

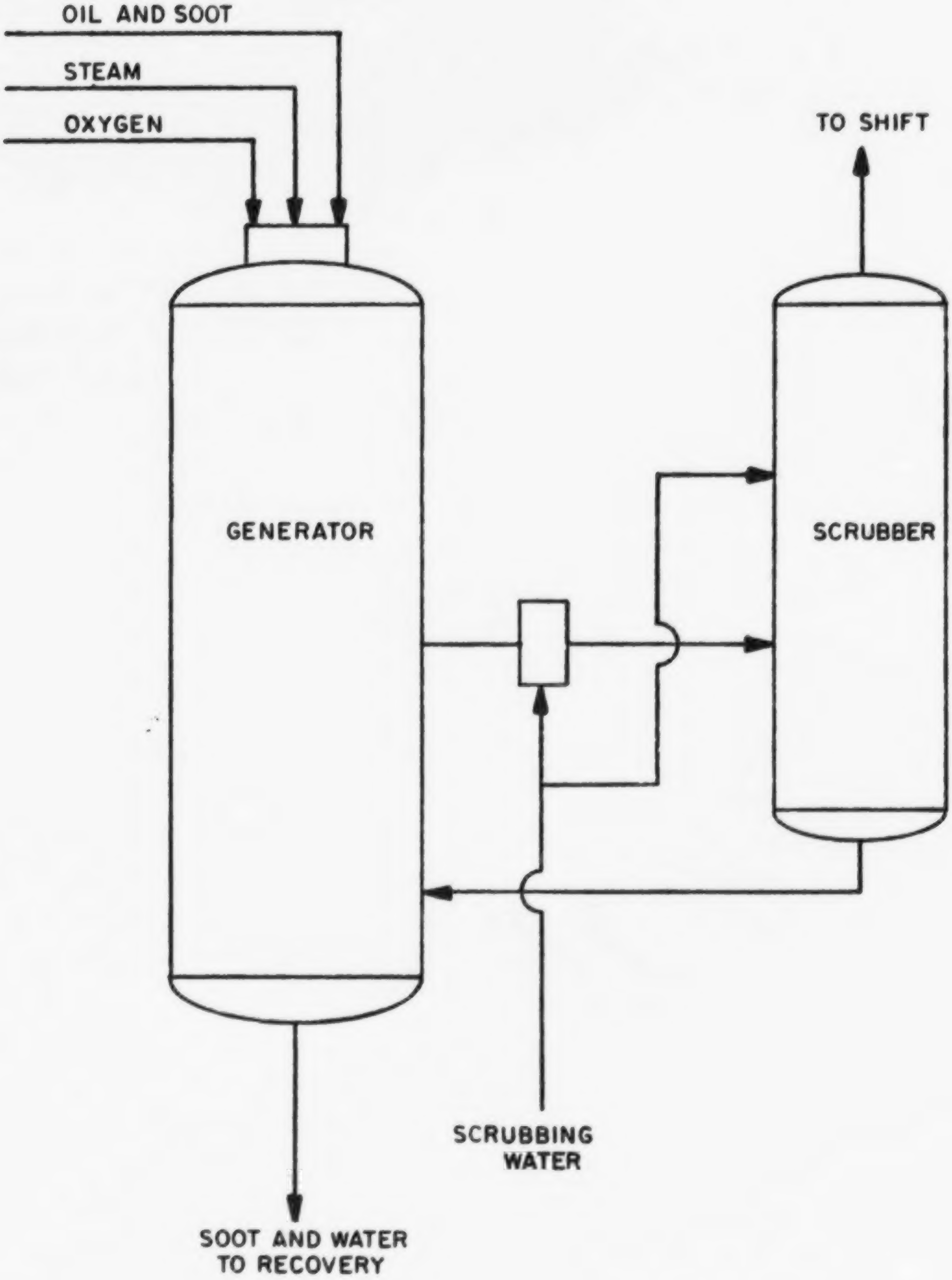
Sulfur in the residuum is converted to hydrogen and carbonyl sulfides and can be removed by any of the commercially available processes such as Rectisol, Benfield, etc. Elemental sulfur is the ultimate by-product.

Feed stocks with up to one weight percent ash have been used commercially without difficulty. Ash components are partially sequestered in ungasified soot, which is scrubbed from the gas with water. The ungasified carbon is ultimately recycled to extinction.

Clean low-Btu (about 130 Btu/SCF) gas is produced if air is the oxidant. The heating value is increased to about 320 Btu/SCF if oxygen is used in place of air.

SNG can be produced if the product gas is shift converted and catalytically methanated.

A wide variety of sulfur-containing distillate and residual fuel oils have been processed commercially.



GENERATOR WITH DIRECT WATER QUENCH

FIGURE 5.9.2. TEXACO PARTIAL OXIDATION PROCESS(1)

TABLE 5.9.2 TEXACO PARTIAL OXIDATION: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
			The Texaco Process has been licensed for use in more than 60 plants in over 20 countries throughout the world for production of high purity hydrogen or synthesis gas. A wide variety of feed stocks have been employed, and 35, primarily the more recent ones, have used heavy oils.

References:
(1) Child, Edward T., "Texaco: Heavy Oil Gasification", presented at the University of Pittsburgh School of Engineering Symposium on Coal Gasification and Liquifaction, Pittsburgh, Pa. (August 6-8, 1974).
(2) Schlinger, W. G. and Slater, W. L., "Partial Oxidation-A Minimum Pollution Route for Hydrogen Manufacture", American Chemical Society Meeting, Petroleum Division, Los Angeles, Calif. (April 1971).
(3) Crouch, W. G., Schlinger, W. G., Klapatch and Vitti, G. E., "Recent Experimental Results on Gasification Combustion of Low-Btu Gas for Gas Turbines", Combustion, pp 32-25 (April 1974).

5.9.3 Shell Gasification Process (Shell Oil Company)

Description of Process. A simplified flow diagram is shown in Figure 5.9.3. The hydrocarbon feed and oxidant (air and/or oxygen) are preheated and fed to the reactor. When the oxidant is either oxygen or enriched air, steam must be injected as well in order to moderate reactor temperature. The principal reaction is parted oxidation of the hydrocarbon to carbon monoxide and hydrogen, but other reactions such as steam cracking, hydrocracking and carbonization also occur.

The hot reactor-effluent (2,200 to 2,400 F), containing about three percent of the feed as soot, is cooled by passage through a waste-heat boiler. Soot is recovered from the crude gas leaving the waste-heat boiler and recycled to extinction with fresh feed. The product gas is virtually free of entrained carbon ($<$ 5 pmm).

Sulfur is converted primarily to hydrogen sulfide and traces of carbonyl sulfide which are removed, together with most of the carbon dioxide, in a Shell Sulfinol process unit. The desulfurized gas typically contains less than 5 ppm sulfur. The ultimate by-product is elemental sulfur.

If air is used as the oxidant, the heating value of the gas is about 120 Btu/SCF. If oxygen is employed the value is about 300 Btu/SCF. Less than one (vol) percent methane is produced.

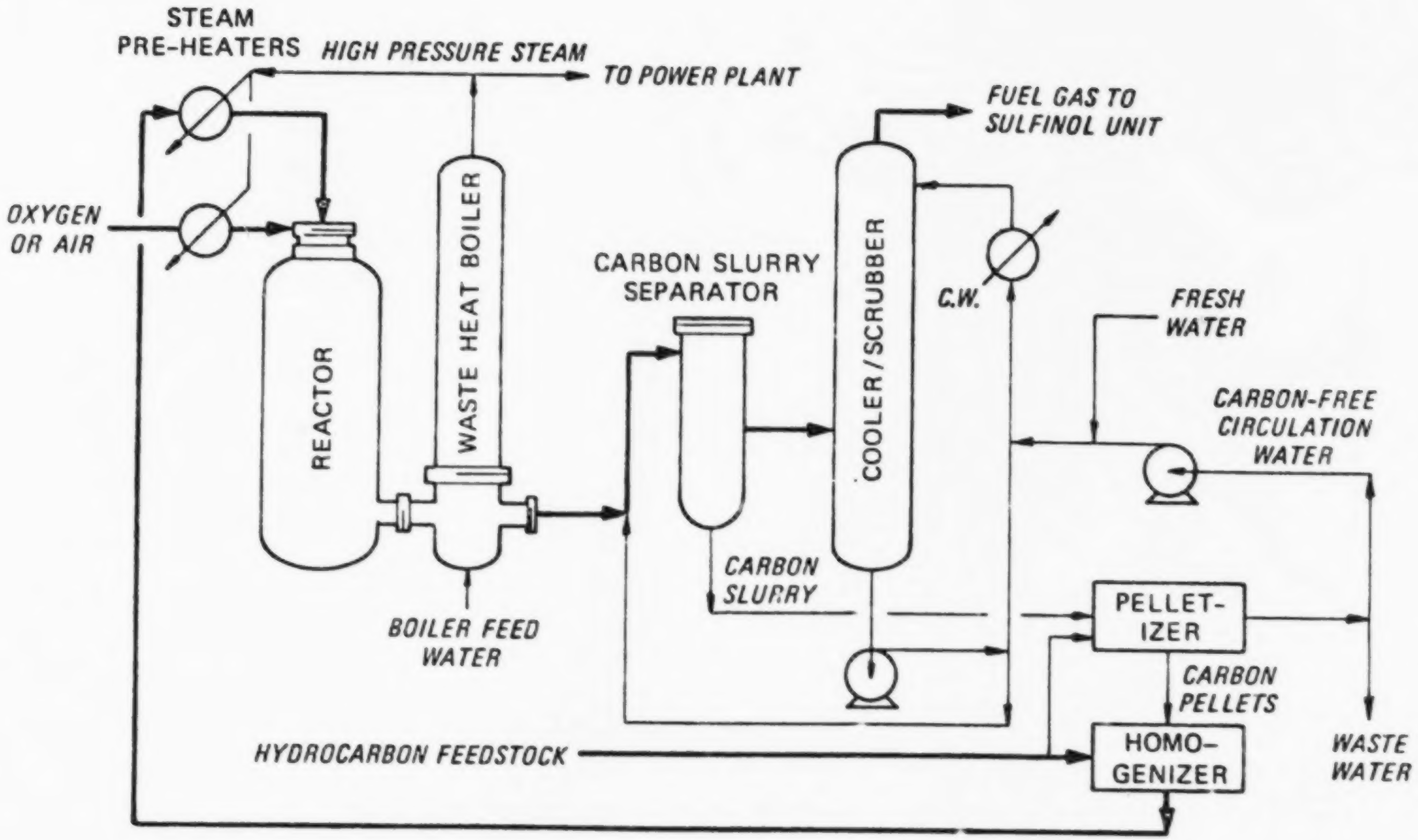


FIGURE 5.9.3. SGP FOR FUEL GAS MANUFACTURE(1)

TABLE 5.9.3. SHELL GASIFICATION PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Commercial		Shell Oil Company	100 reactor units have been installed in 34 plants around the world since 1958. They operate on a variety of feedstocks. Largest plant (Germany) produces 140-160 MMSCF/D of synthesis gas for production of methanol and ammonia.
(Contact: J. A. Sykes, Jr., Shell Oil Company, Houston, Texas (713) 795-3903)			

References:
(1) Dravid, A. N., Kuhre, C. J., and Sykes, J. A., Jr., "Power Generation Using the Shell Gasification Process", Presented at the Third International Conference on Fluidized-Bed Combustion (Oct. 1972).
(2) Plummer, J. B., Kuhre, C. J., Reed, C. L., and Sykes, J. A., Jr.,"The Generation of Clean Gaseous Fuels from Petroleum Residues," Presented at the American Institute of Chemical Engineers Meeting, Tulsa, Oklahoma (May 11-13, 1974).
(3) Kuhre, C. J., and Sykes, J. A., Jr., "The Shell Gasification Process for the Substitute Natural Gas Industry", Presented at the IGT-SNG Symposium, Chicago (March 12-16, 1973).

5.9.4 H-Gas (Hydrocarbons Research Inc)

Description of Process. A simplified flow diagram is shown in Figure 5.9.4. Hydrogasification and gasification steps are carried out in separate zones within a single reactor vessel. Feedstock is hydrogasified to produce light hydrocarbon gases. Coke and some heavy residual oil are gasified with steam and oxygen to generate hydrogen for the hydrogasification.

Coke generated in the hydrogasification zone passes to the gasification zone on an inert heat carrier. The coke is gasified, and the inert carrier passes back to the hydrogasification zone.

Gases from the hydrogasifier are quenched with a stream of gas oil and fed to a liquid recovery section. Liquid hydrocarbons (C_5 and higher) are recovered by fractionation and absorption. Recovered heavy gas oil is fed to the gasification zone. A portion of light distillate is recycled to the hydrogasifier zone of the reactor.

The net yield of C_5 - 400 F distillate is fed to a hydrotreater for desulfurization. The product from this process provides the fuel required by the plant.

Condensed water, containing dissolved ammonia and acid gases, is recovered and fed to the water-treating system.

Figure 5.9.4 shows the additional processes required to upgrade the gas to SNG. In this case, the gaseous product stream from the liquid recovery system is fed with high-pressure steam to a shift converter. The effluent from the shift converter is treated to remove acid gases and then is catalytically methanated to SNG.

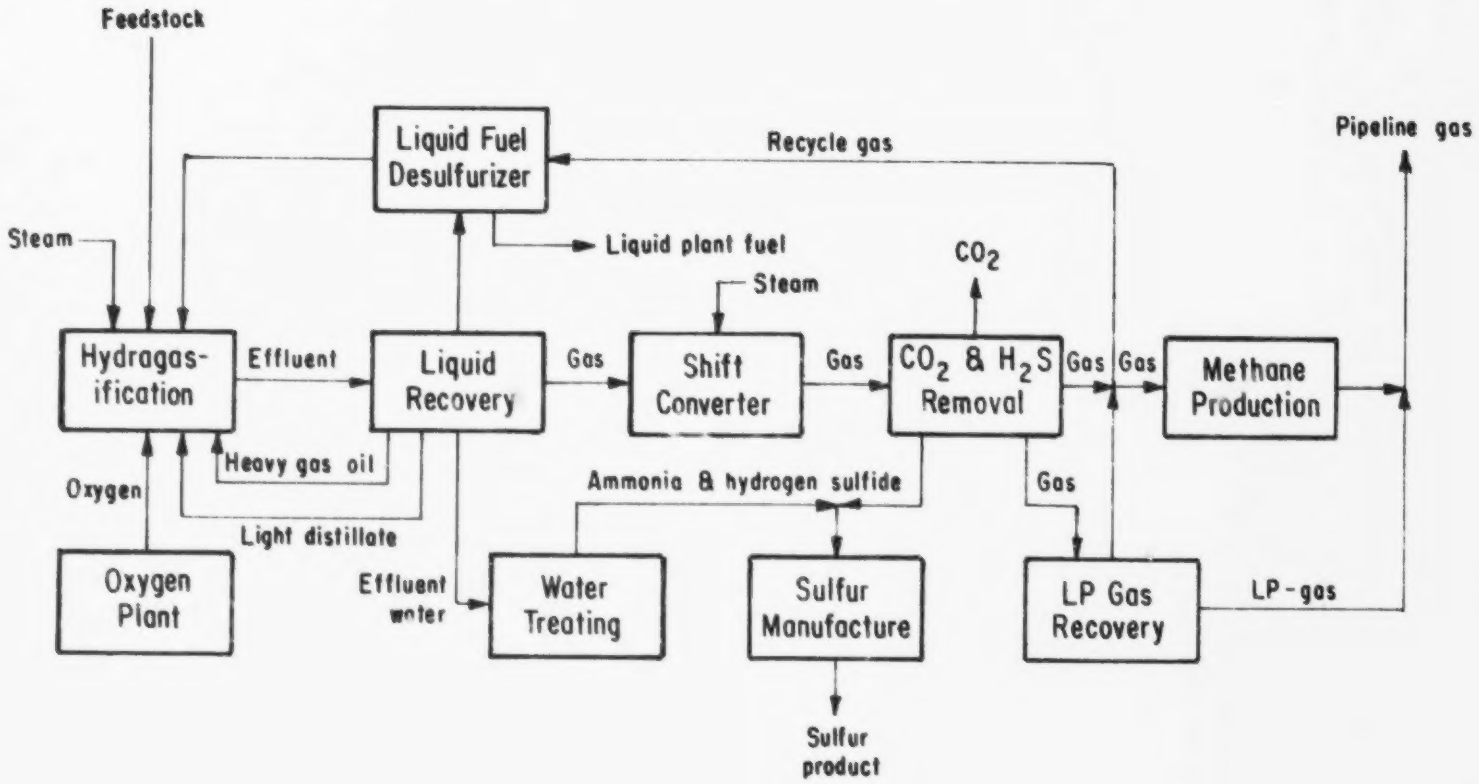


FIGURE 5.9.4. H-GAS PROCESS(1)

TABLE 5.9.4. H-GAS: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
		Hydrocarbon Research Inc.	Bench-scale studies with a unit capable of processing up to 15 B/D have been completed. Results demonstrate that hydrogasification process is applicable to processing resid, heavy sour crudes and heavy sour distillate fractions.
	(Contact: Jim Livingston, Hydrocarbons Research Inc, New York, N.Y. (212) 349-1480		

Reference:
(1) Anon, "SNG Process Passes Pilot Plant Test", The Oil and Gas Journal, pp 32-33 (April 9, 1973).

5.9.5 IGT (Institute of Gas Technology)

Description of Process. A schematic diagram of the process is shown in Figure 5.9.5. Feedstock is hydrogasified at 1,300 to 1,400 F and 1,000 psi in a fluidized bed of solids. The solids act as a heat carrier and also carry off excess carbon.

Carbon-coated solids from the hydrogasifier are continuously circulated to a second vessel where the carbon is gasified at 1700-1800 F and 1000 psi with steam and oxygen to produce hydrogen for the hydrogasifier. The high pressure leads to significant yields of methane directly from the residue carbon. The raw gas is shift converted and treated for acid-gas removal before being sent to the hydrogasifier.

Raw gas from the hydrogasifier is cooled and scrubbed to remove excess steam and light oils and then desulfurized. The product is not expected to require catalytic methanation. The heating value should be about 930 Btu/SCF.

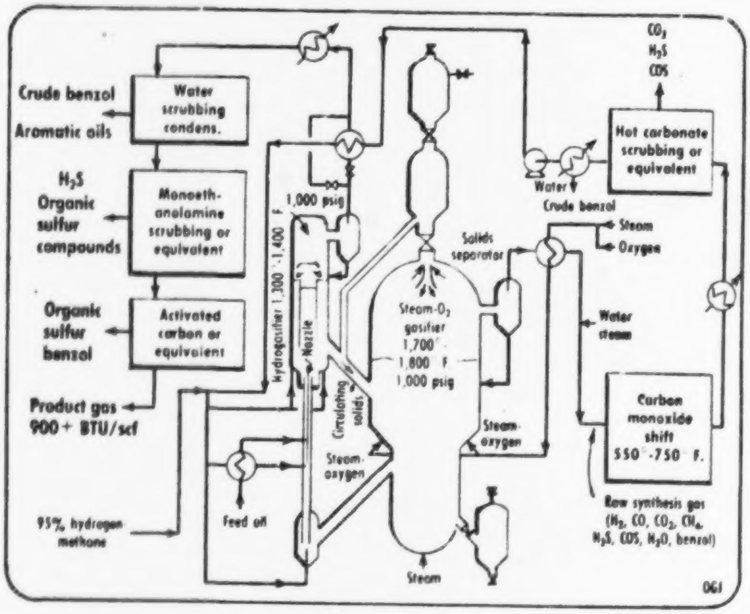

Crude benzol
Aromatic oils
Water scrubbing condens.
H_2S
Organic sulfur compounds
Monoethanolamine scrubbing or equivalent
Organic sulfur benzol
Activated carbon or equivalent
Product gas 900+ BTU/scf
95% hydrogen methane
Feed oil
1,000 psig
Hydrogasifier 1,300°-1,400° F
Nozzle
Circulating solids
Steam-oxygen
Solids separator
Steam-O_2 gasifier 1,700°-1,800° F 1,000 psig
Steam
Steam-oxygen
Water
Crude benzol
Steam
Oxygen
Water steam
CO_2
H_2S
COS
Hot carbonate scrubbing or equivalent
Carbon monoxide shift 550°-750° F.
Raw synthesis gas (H_2, CO, CO_2, CH_4, H_2S, COS, H_2O, benzol)
OGI

FIGURE 5.9.5. THE IGT PROCESS

TABLE 5.9.5. IGT: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
		IGT	Laboratory studies of the kinetics of oil char gasification. Economic analysis.

(Contact: Dennis Duncan, IGT, Chicago, Illinois (312) 225-1455)

References:
(1) Anon, "High-Sulfur Resid Eyed for SNG Feed", Oil and Gas Journal, pp 36-37 (February 12, 1973).

SECTION 6

HIGH-BTU GAS FROM COAL

Introduction

Between 1946 and 1971 natural gas production increased 351 percent while reserves increased by only 90 percent*. Since the American Gas Association (AGA) began publishing natural gas statistics in 1946, new additions to reserves exceeded production every year until 1968. In that year the situation was reversed and that trend has continued in subsequent years with the gap between production and addition to reserves growing increasingly wider**.

The Federal Power Commission has estimated that the annual deficit in domestic natural gas may reach 17.1 trillion cubic feet by 1990***. Clearly, a number of alternative sources must be developed if the projected gap between supply and demand is to be bridged.

The production of high-Btu gas from coal probably has the greatest long-term potential for assuring an adequate supply of pipeline-quality gas to meet our national requirements. High-Btu gas from coal is more than 90 percent methane and has a heating value in the range 900 to 1,000 Btu/scf. It is prepared by upgrading the mixture of carbon monoxide, carbon dioxide, hydrogen and methane generated by the primary gasification of coal. Upgrading includes purification, increase in the ratio of hydrogen to carbon monoxide by the water-gas shift reaction and catalytic methanation to convert carbon monoxide and hydrogen to methane (Figure 6.0).

References for a given process are given at the foot of the respective "State of the Art" table. General references for coal gasification are given on Page 6-37.

General environmental factors associated with the gasification of coal are discussed in Appendix A. Because of the considerable overlap in the environmental aspects of producing the various qualities (in Btu/SCF) of gas from coal all of the processes described in this section and in Section 5 (low- and intermediate-Btu gas) are treated together in the discussion in Appendix A.

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* Federal Power Commission, Annual Report (1972).

** Except for 1970, in that year, addition to reserves exceeded production because of the inclusion of the vast but remote North Slope reserves in Alaska.

*** National Gas Supply and Demand 1971-1990, FPC Staff Report No. 2 (Feb., 1972).

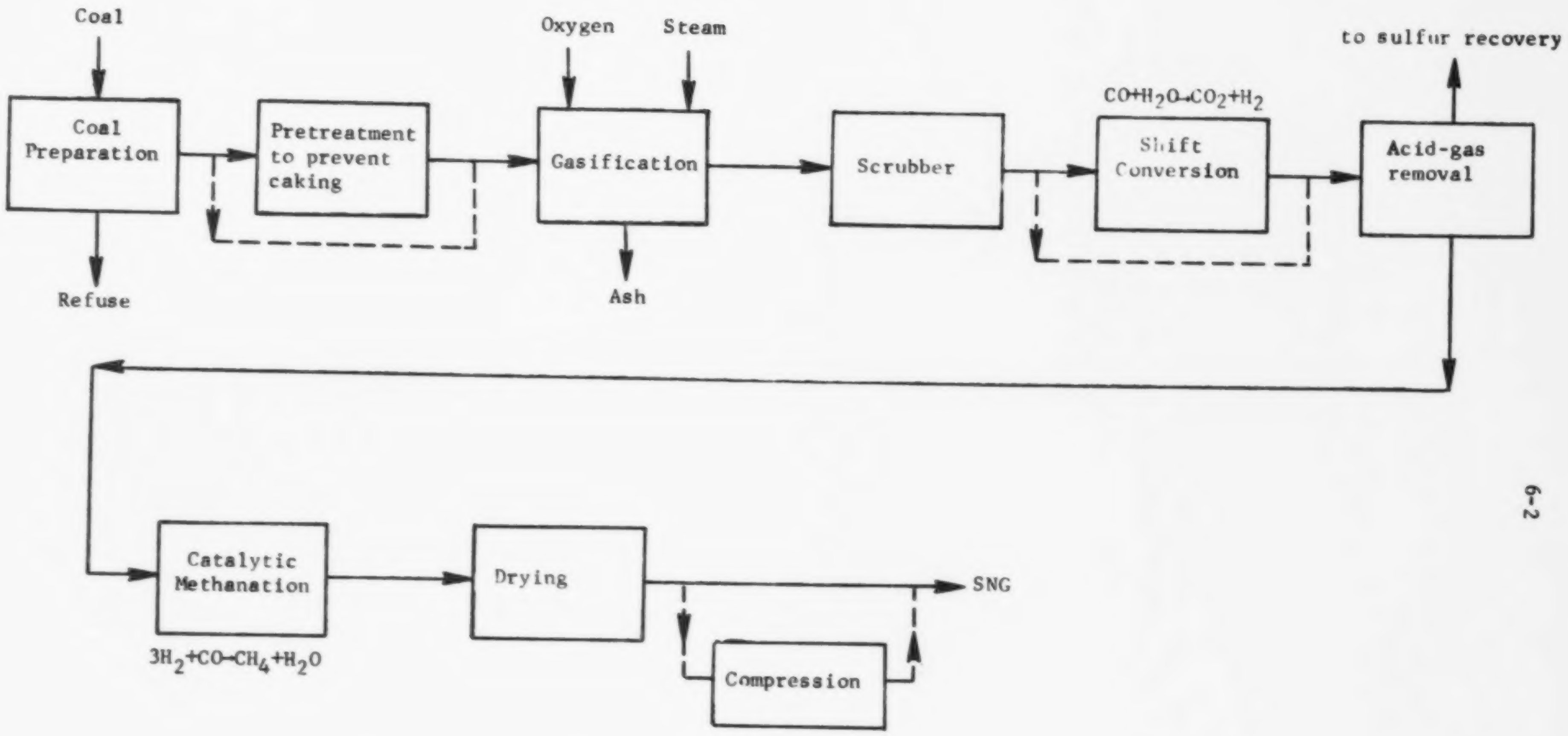

FIGURE 6.0. GENERALIZED SCHEMATIC FOR PRODUCTION OF SNG FROM COAL

Status of the Technology

Applications have already been filed for two commercial-scale (250 MMSCF/D) plants for the production of SNG from coal. Both will be based on the Lurgi process, and construction of the first of these plants is expected to begin in the third quarter of 1975, with start-up estimated for early 1978. At least two other plants of this size, also based on Lurgi technology, are in the planning stage (see Table 6.1).

Plants based on emerging technology are expected to result in lower capital investment and operating costs -- and hence in lower SNG prices. However, none of the new processes is beyond the pilot plant stage (Table 6.0), and it is unlikely that commercial-scale plants based on any of these will be built before 1980.

All of the plants presently in the pilot stage have received support from the Office of Coal Research (OCR), the American Gas Association or both. Funding of a number of alternative processes is desirable because of the rather considerable extension of technology involved and the consequent danger of encountering "dead-ends".

Hopefully, at least one of the processes now under development will show sufficient promise to warrant the design and construction of a demonstration plant to determine commercial feasibility. This aspect of the development program will probably require generous government support. OCR has proposed that $1.5 billion will be required for design, site selection, construction and operation of five demonstration plants (40), but that figure is probably too low.

At this stage of development, it is difficult to anticipate potential problem areas. An evaluation by the National Academy of Engineering (7) identified some of the major advantages and disadvantages of several of the new processes and outlined specific problems associated with the more advanced of these. Hottel and Howard (1) made a rather similar analysis a few years earlier. In Tables 6.1-6.12 that follow, the state-of-the-art

TABLE 6.0. STATUS OF HIGH-BTU GASIFICATION: A SUMMARY

Process/Developer	Current Status	Comments
Lurgi	Demonstration tests (up to 2.5 MM SCF/D) of production of SNG from coal have been carried out in Westfield, Scotland. Applications have been filed with FPC for construction and operation of two 250 MMSCF/D plants in New Mexico (see Table 6.1).	Major disadvantage is small throughput, size of gasifier is limited. Commercial scale plants will require several (27-33) gasifiers in parallel. As a result, capital investment and operating costs are relatively high. Also, relatively low yield of CH_4 (<10%) during gasification. Results at Westfield and Sasolburgh (Table 6.1) indicate methanation is now commercially feasible.
Koppers-Totzek	Gasifier technology is well-developed (commercial). No plans have yet been announced to use K-T gasifiers in an SNG-plant.	Major disadvantage is the very low yield of CH_4 (~0) during gasification.
Hygas/IGT*	3TPH pilot in operation in Chicago since May 1971.	Plant includes all essential operations except shift conversion. The most advanced of the new processes. The complexity of the process-in particular transfer and injection of char at high temperature and pressure-is a problem. Mid-1975 target date for design of a commercial plant.
Steam Oxygen	Present pilot is integrated with a steam-oxygen unit for producing hydrogen for the gasifier.	Presently undergoing shakedown.
Steam Iron	Blaw-Knox is presently designing a steam-iron unit.	May prove to be most economical way to produce hydrogen for the Hygas process.
Electrothermal	Built and tested briefly but presently "mothballed"	Considered to be too expensive at present.
Synthane*/US Bureau of Mines	3 TPH pilot plant is under construction at Bruceton, Pa.	Construction is about 60% completed as of July 1974. Expect completion in January 1975. This will be a fully integrated plant. Relatively simple process. The relatively low yield of CH_4 (25-30%)** during gasification is a disadvantage.
Bigas*/Bituminous Coal Research	5 TPH pilot plant is under construction at Homer City, Pa.	Expect completion in early 1975. Plant will be fully integrated. Throughput for this process will be high but a large O_2 plant is required and the relatively low yield of CH_4 (20%)** during gasification is a disadvantage.
CO_2Acceptor*/Consolidation Coal	1.5 TPH pilot plant in operation since April 1972 at Rapid City, S.D.	Plant does not include provisions for methanation. Many of the problems that plagued earlier runs have been solved and operations are now going more smoothly.

TABLE 6.0 (Continued)

Process/Developer	Current Status	Comments
		Major advantage is that acceptor eliminates need for O_2 plant and reduces the need for shift conversion. However, a very complex fluidized system is involved and control and scale-up may present problems. Other disadvantages include a rather low yield of CH_4 (~15%)** during gasification and the fact that only lignite and sub-bituminous coals can be processed.
Hydrane*/US Bureau of Mines	1 TPH pilot is presently being designed.	Work thus far has been limited to laboratory-scale (see Table 6.7). The major advantage of the process is the high yield (~75%)** of CH_4 from hydrogasification. However, this is a relatively complex operation and may require extensive development.
Ash Agglomeration*/ Battelle, Union-Carbide	1200 lb/hr PDU is presently under construction at West Jefferson,O	Estimate completion in first quarter of 1975. Gas purification, shift conversion and methanation are not included. Major advantages are that no O_2 plant is required and that a relatively clean product gas is obtained. Disadvantages include the low CH_4 yield (less than 5%)** and the difficulty of transferring hot ash agglomerates between combustor and gasifier.
Kellogg Molten Salt/ M.W. Kellogg Co.	Bench scale.	Molten carbonate creates serious corrosion problems; these apparently have been solved by use of Monofrax A.Process is rather complex. Direct yield of CH_4 is low (~5%)**. A process development unit is planned (preliminary flow sheets and cost estimates have been prepared).
Atgas (Molten Iron)/ Applied Technology Corp.)	Bench scale.	Lance design is complicated (must be compatible with molten iron and slag). Essentially no CH_4 is formed in the gasifier because of the low pressure and high temperature.
Garret Flash Pyrolysis/ Garret Research and Development	Bench Scale (50 lb/hr) in operation since January 1973.	A 10 TPH pilot plant has been proposed. Process produces large quantities of tar and char as byproducts.
COGAS/Cogas Development Co.	4 TPH pilot in operation since March 1974 in Leatherhead, England. Also, a 400 lb/hr pilot in operation since May 1974 in Plainsboro, N.J.	Expect test runs to cover an extended duration and provide basis for construction of a larger pilot or a demonstration plant.

*Presently funded by OCR or US Bureau of Mines.

**Methane analysis in volume % on a dry, CO_2-free basis. Values are only approximate and will depend upon the coal being gasified.

for each of twelve new processes is reviewed, and Table 6.0 presents a summary view of the current status of these processes with some comments on problem areas.

At the same time, it is useful to consider some of the more general developmental problems associated with unit operations common to most processes.

Coal Mining. The development of advanced surface and underground mining technology that will increase productivity and coal recovery and at the same time meet health, safety and environmental standards will have an important bearing on coal gasification.

Coal Preparation. At present it is not possible to crush or grind coal to a specific size without production of surplus fines. In most of the fluid-bed processes the coal must be sized to prevent high fuel losses by carbon-carryover, and most fixed bed processes cannot accept fines.

Coal Feeding. Because most of the new processes operate at high pressures, coal feeding is a major design problem. Lock-hoppers are most commonly used, but they result in rather large energy losses, and it is difficult to find a material that will seal against high pressures after more than a relatively few cycles. Other feed systems such as slurries and piston feeders are being investigated, but considerably more development work is required.

Refractory Problems. Under conditions prevailing in the gasifier SiO_2 reacts with hydrogen to produce SiO and water vapor. This reaction is reversed downstream, where temperatures are lower, and pipes can become clogged with SiO_2. Presently, the approach is to use expensive alumina refractories (39).

Hot Char Transport and Injection. Many of the new processes require the recycling of char. Development of a system capable of transporting and injecting char at high temperatures and pressures without producing serious erosion is a problem which requires much more work.

Slag and/or Ash Removal. The high operating pressures involved in most of the new processes make the withdrawal of slag or ash from the gasifier a difficult design problem. The most commonly proposed method involves quenching in water and removal as a slurry through a lock-hopper system.

Gas Clean-up. Incomplete removal of char from the gasifier effluent could result in poisoning of the shift catalyst or in char accumulation downstream.

Quench Chamber Corrosion. In many cases, the gasifier effluent is quenched to scrub out heavy hydrocarbons, and particulates. Because of the high pressures and temperatures and the corrosive nature of the gas (relatively high concentration of hydrogen sulfide), materials for this stage of the system are subject to severe corrosion. Considerable research is still needed on materials for this stage (39).

Methanation. The recent studies by CONCO Methanation Company at Westfield Scotland and by Lurgi in Sasolburg, South Africa (see Table 6.1) are encouraging. In the Westfield demonstration, up to 2.5 x 10^6 SCF/day of SNG were produced by methanation of gas from a Lurgi gasifier after shift conversion and purification. Technical details have not yet been published.

In any case, the development of new and imporoved catalysts and reactors for methanation should continue to be an area of considerable activity.

Char Utilization. Char is produced in some of the advanced gasification process. Some of this is expected to be used to generate hydrogen and/or process steam. Thus far, however, these processes for char utilization have received rather limited attention.

The AGA recently identified 176 sites with sufficient uncommitted reserves of coal mineable at acceptable costs and with adequate water to operate SNG plants, each capable of producing 250×10^6 SCF/day, over a period of at least 25 years. 141 of these sites are west of the Mississippi River.

A developing coal gasification industry will require the continued support of strong research programs, both fundamental and applied, in such areas as reaction kinetics, catalysis, gas purification, materials, etc.

Over the longer range, it is probable that new methods will continue to emerge with the potential for lower SNG costs and more efficient utilization of coal. It is important that funding agencies and the industry itself remain sufficiently alert and flexible to encourage and promote promising new approaches.

6.1 The Lurgi Process (American Lurgi Corp.)

Description of the Process. The primary gasification process is described in Section 5.1. To prepare pipeline-quality gas, the raw gas from the Lurgi gasifier is shift-converted to increase the hydrogen/carbon monoxide ratio, purified and catalytically methanated.

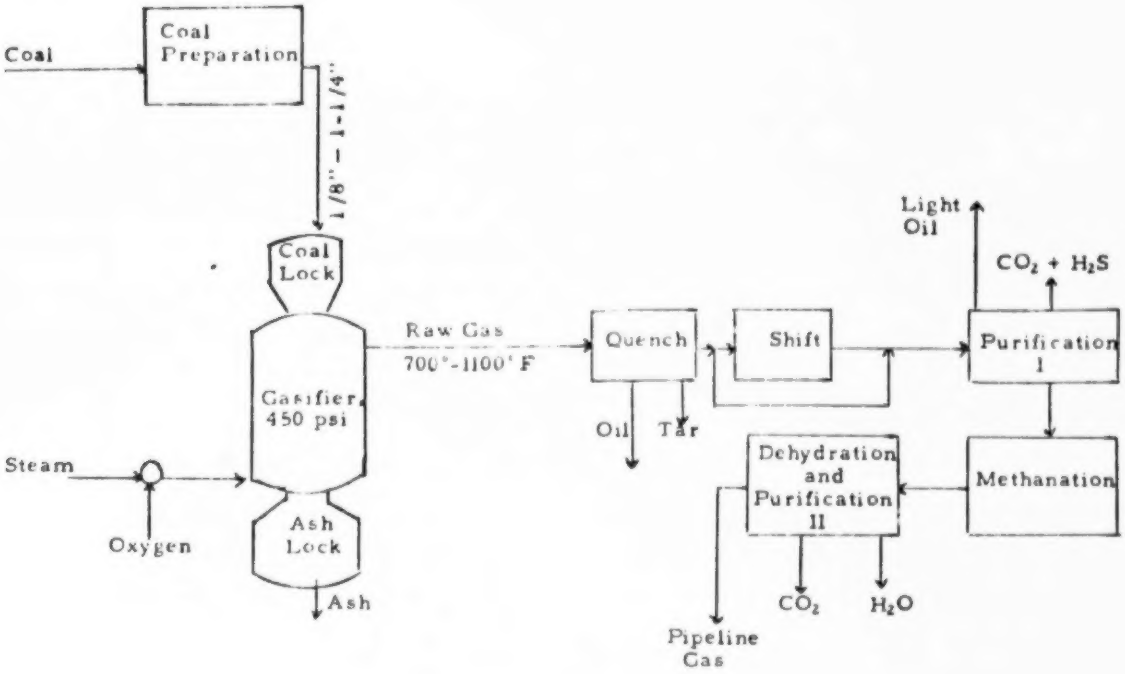

FIGURE 6.1. LURGI PROCESS

TABLE 6.1 LURGI: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Methanation pilot	Sasolburgh, South Africa	South African Coal, Oil and Gas Corp.	In conjunction with Lurgi Mineraloeltechnik GmbH, SASOL has studied catalytic methanation of CO-rich synthesis gas--a slip stream of about 700 scf/hr from SASOL's commercial plant (Table 5.1.1)--with a special methanation catalyst developed by BASF. Results during the 1-1/2 yr test indicate methanation can be carried out without carbon formation to yield an SNG with less than 1% (Vol.) H_2 and less than 0.1% CO and a heating value of 970 Btu/scf.[2] Trace components in synthesis gas leaving Rectisol wash have little influence on catalyst activity and life. Based on results for runs of 2600 hrs they calculate an expected life time of 16,000 hrs for the special BASF catalyst(6,7).
Methanation pilot	Westfield, Scotland	Sponsored by a group of U.S. oil companies headed by Continental Oil Co in collaboration with the Scottish Gas Board	Town gas from the Westfield plant (Table 5.1.1) is being upgraded to SNG as a demonstration of commercial feasibility. Up to 2.5 MMSCF/D of SNG is produced by methanation of purified (Rectisol) gas in a fixed-bed adiabatic reactor containing a Ni-based catalyst. Product gas is 90-95% CH_4 and has a heating value of about 980 Btu/SCF(8). Results demonstrate that methanation is commercially feasible. Tests will conclude in September 1974.
50×10^6 SCF/day (Demonstration)	Burnham, N.M.	ElPaso Natural Gas (plant will be owned and operated by Fuel Conversion Co. - a wholly owned subsidiary)	Awaiting final FPC approval. Expect to begin construction in 3rd quarter of 1975 and to begin operation in mid-1976. Capital investment will be $\$20 \times 10^6$. Function will be to test process operation. Goals include operating at 20% above design capacity and 30% above design pressure. Will also be used to train operators for their 250 MMSCF/D facility and the Wesco plant (see below).
250×10^6 SCF/day	"	"	Construction will begin concurrently with the test facility described above. Expect start-up in Jan. 1978 and full production in July 1978. Gas will cost \$1.51/103 ft^3 (first year price in 1973 dollars). Capital investment in plant and mine is estimated at $\$605 \times 10^6$.

TABLE 6.1 LURGI: STATE OF THE ART (CONTINUED)

Facilty	Location	Owner(s) or Contractor	Status/Operating History
250 x 10^6 SCF/day	San Juan County, N.M.	Pacific Coal Gasification Co., and Transwestern Coal Gasification Co. (WESCO will build and operate the plant; Utah International Inc. will supply water and coal.)	Authorization for the project filed with FPC on Feb., 7, 1973. Capital costs estimated to be about \$500 x 10^6 Slated for completion in late 1977.
250 x 10^6 SCF/day	Eastern Wyo.	Panhandle Eastern Pipeline Co. and Peabody Coal Co.	Feasibility study in progress (by M. W. Kellogg and American Lurgi Corp.). Projected for operation in 1978-80.
N.A.	Southern Illinois	"	Feasibility studies are underway.
1000 x 10^6 SCF/day (4 plants)	Dunn County, North Dakota	Natural Gas Pipeline Company of America	Plans are for four plants and possibly eight. Mining will be from 110,000 acres of leased land in N. Dakota. Each mine will cost about \$100 million and the associated gasification plant \$370 million. The first plant is scheduled to go on line in 1982. (9)

References:

(1) Hottel, H.C. and Howard J.B., New Energy Technology; Some Facts and Assessments, MIT Press, Cambridge, Mass. (1971).

(2) The Supply-Technical Advisory Task Force-Synthetic Gas-Coal, prepared by the Synthetic Gas-Coal Task Force for the Federal Power Commission (April 1973).

(3) Rudolph, Paul F. N., "The Lurgi Process, The Route to SNG from Coal", presented at the Fourth Snythetic Pipeline Gas Symposium, Chicago (1972).

(4) "Evaluation of Coal Gasification Technology. Part I. Pipeline Quality Gas", R&D Report No. 74-Interim Report No. 1, Office of Coal Research, Washington, D.C. (1973).

(5) Moe, James M., "SNG from Coal via the Lurgi Process", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 91-110, Chicago (1973).

TABLE 6.1 (Continued)

Facilty	Location	Owner(s) or Contractor	Status/Operating History

References Continued:

(6) Moeller, F. W., Roberts H., and Britz B., "Methanation of Coal Gas for SNG", Hydrocarbon Processing. pp 69-74 (April 1974).

(7) Hoogendoorn, Jan C., "Gas from Coal with Lurgi Gasification of SASOL", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 91-110, Chicago (1973).

(8) Anon, "Coal Gasification Plant Begins Operation", Chemical and Engineering News, p 21 (November 5, 1973).

(9) "Report to Project Independence Blueprint, Federal Energy Agency", Prepared by the Interagency Synthetic Fuels Task Force, Supplement 1 (July 8, 1974).

6.2 The Koppers-Totzek Process (Heinrich Koppers, G.m.b.H)

Description of the Process. The primary gasification process is described in Section 5.6.1. The raw gas from the Koppers-Totzek gasifier is desulfurized, shift converted and catalytically methanated.

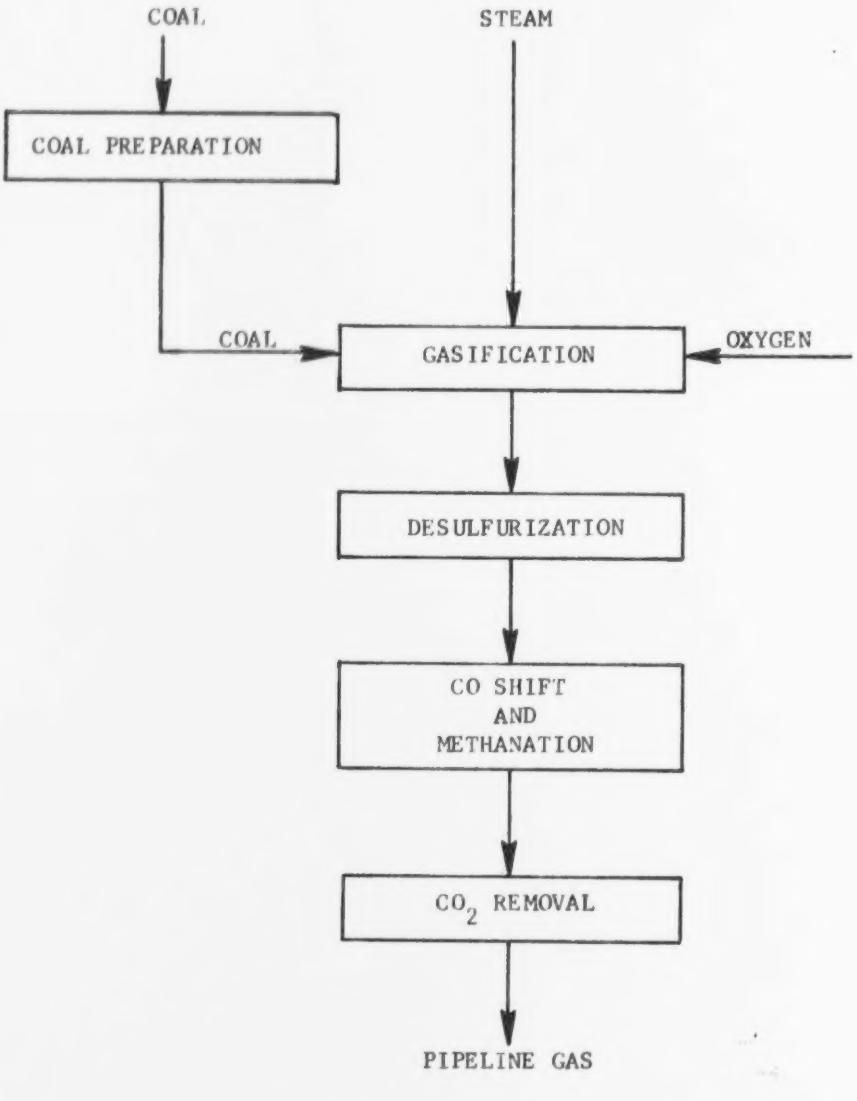

FIGURE 6.2. KOPPERS-TOTZEK(1)

TABLE 6.2. KOPPERS-TOTZEK: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
			The Koppers-Totzek gasifier has been used on a commercial scale for production of synthesis gas for a number of years (see Table 5.6.1). However, it has not been used to prepare SNG. Cost estimates of SNG production have been made (1) and potential environmental effects have been considered (2).

(Contact: J. F. Farnsworth, Koppers Engineering and Construction, Pittsburgh, Pa. (412) 391-3300)

References:
(1) Farnsworth, J. Frank, Mitsak, D. M., Leonard, H. F., and Wintrell, Reginald, " Production of Gas from Coal by the Koppers-Totzek Process", IGT Symposium on Clean Fuels from Coal, pp 143-162, Chicago (1973).
(2) Magee, E. M., and Shaw, H., "Evaluation of Pollution Control in Fossil Fuel Conversion Processes, Koppers-Totzek Process", prepared for Office of Research and Development, USEPA, Washington, D.C. (January 1974).

6.3 The Hygas Process (Institute of Gas Technology)

Description of the Process. There are three Hygas processes (Figure 6.3) which differ essentially only in the method of generating the hydrogen required for the fluidized-bed hydrogasifier.

Coal is crushed, dried and sized, and, if necessary, treated with air at 750-800 F to prevent caking. It is then slurried with light oil (a by product of the process) and pumped into the upper section of the hydrogasifier at 600 F and 1000 psig, where most of the oil is evaporated and recovered for recycle.

In the next stage the coal is devolatilized and partially methanated at 1200-1400 F by hot, hydrogen-rich gas. Devolatilized char passes into the bottom stage where it is partially gasified at 1700 F by reaction with steam and hydrogen-rich gas. Residual char which still contains unreacted carbon can be used in the processes described below to generate the hydrogen-rich gas for the hydrogasifier.

Raw gas from the hydrogasifier is purified, passed through a shift converter, and then catalytically methanated.

Variations of the Hygas Process

(1) Electrothermal. Residual char from the hydrogasifier is reacted with steam to produce a hydrogen-rich gas. The heat required for this highly endothermic reaction is furnished by direct-current heating of a fluidized bed of char. Excess char from this electrothermal reactor is used to generate the electrical power required.

(2) Steam-Iron. In this variation of the Hygas process hydrogen for the gasifier is produced by the reaction of steam with iron at 1500 F and 1000 psi. The resulting iron oxide is reduced back to iron by producer gas generated from spent char. This process is potentially superior in efficiency and economics to both the electrothermal and the steam-oxygen processes.

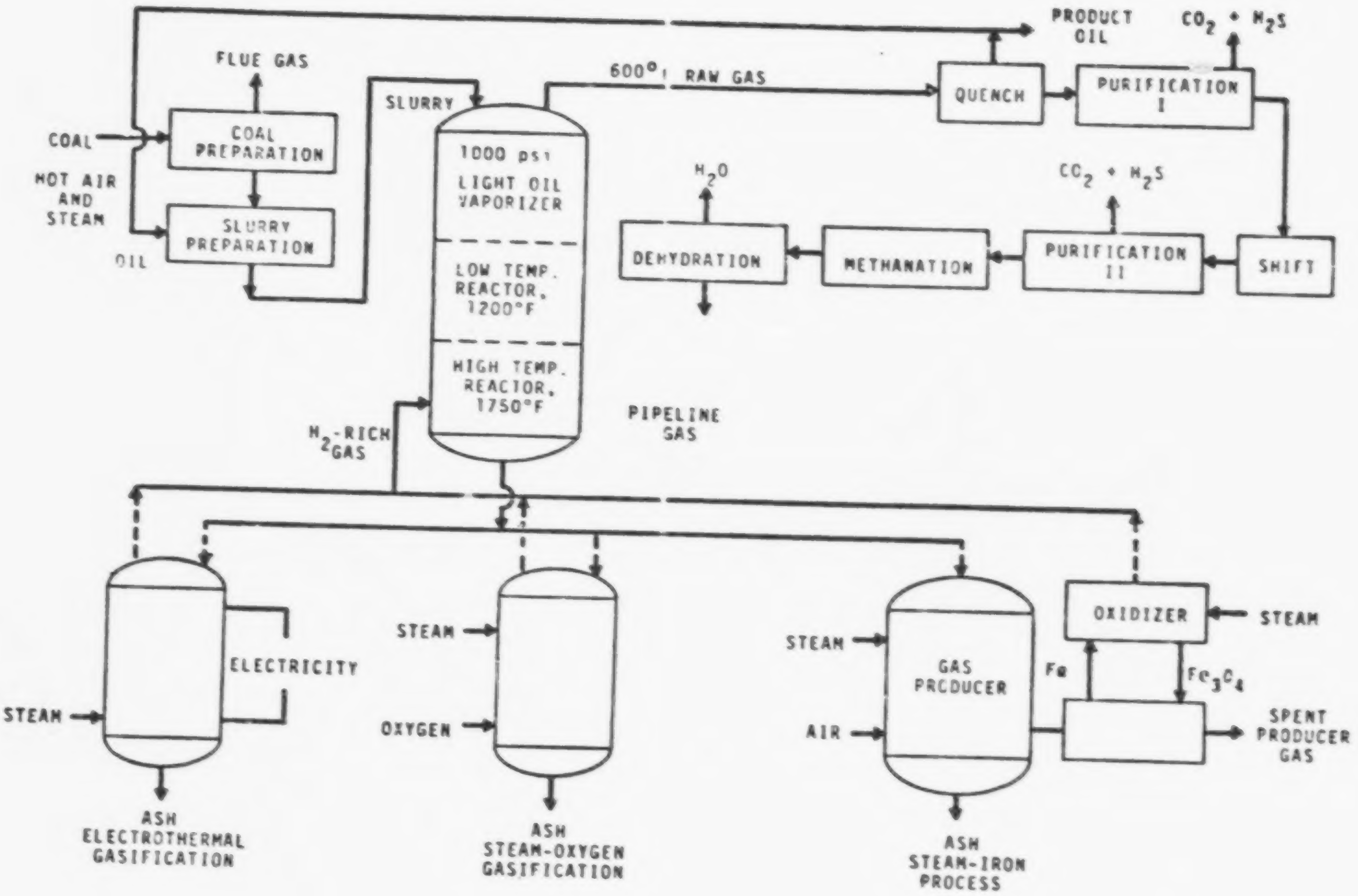

FIGURE 6.3. HYGAS PROCESS(4)

(3) Steam-Oxygen. Similar to electrothermal variation except that the heat required for the char-steam reaction is supplied by combustion of a portion of the char with oxygen. The quantities of steam and oxygen used relative to char are adjusted to maintain an operating temperature of about 1800 to 1900 F.

TABLE 6.3. HYGAS: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
3 ton/hour Pilot	Chicago, Illinois	Institute of Gas Technology (OCR and AGA Funded)	Started operation in May 1971, Cost $\$10^7$. Initially used H_2 produced by reforming CH_4. Pilot does not include shift converted (technology for this process is well developed. Methanation carried out over Ni on keiselguhr). Steam-O_2 plant to produce H_2 for gasifier is now completed and undergoing shakedown (July 74). An electrothermal unit was built and tested briefly but not now economically attractive and unit has been "moth balled" Steam-iron unit is being designed by Blaw-Knox. This may prove to be most economical method of generating H_2 for the process. Operating problems with "off-the-shelf" equipment caused frequent shutdowns earlier. These are being solved and semi-continuous operation of gasifier has been achieved. Operated at pressures up to 1300 psi. Continuous runs of more than 100 hrs have been made with fully integrated unit. Product gas was less than 0.1 ppm S, and had HV of 930-1007 Btu/SCF (N_2-free basis) Longest continuous run to date (July 1974) was 28 days. Mid-1975 target date for design of a commercial scale (250 MMSCFD) plant. Will involve 3 parallel trains.
	(Contact: Bernard S. Lee, Institute of Gas Technology, Chicago, Illinois (312) 542-7080)		

References:

(1) Hottel, H. C., and Howard, J. B., New Energy Technology, Some Facts and Assessments, Cambridge, Mass., MIT Press (1971).

(2) "Evaluation of Coal Gasification Technology. Part I. Pipeline Quality Gas", R&D Report No. 74 - Interim Report No. 1, Office of Coal Research, Washington, D.C. (1973).

(3) The Supply-Technical Advisory Task Force - Synthetic Gas-Coal, prepared by Synthetic Gas-Coal Task Force for the Federal Power Commission (April 1973).

(4) Schora, F. C., Jr., Lee, B. S., Huebler, J., "The Hygas Process", p 219, Institute of Gas Technology Symposium on Clean Fuels from Coal, Chicago (1973).

(5) Annual Report for Calendar Year 1973, OCR, U.S. Dept. of the Interior.

(6) Annual Report 1973-74, OCR, U.S. Dept. of the Interior.

6.4 The Synthane Process (Bureau of Mines)

Description of the Process. The primary gasification is described in Section 5.3.2. Raw gas from the fluid-bed gasifier is cleaned, passed through a shift converter, scrubbed almost free of sulfur compounds and carbon dioxide, and then catalytically methanated to produce pipeline quality gas (Figure 6.4).

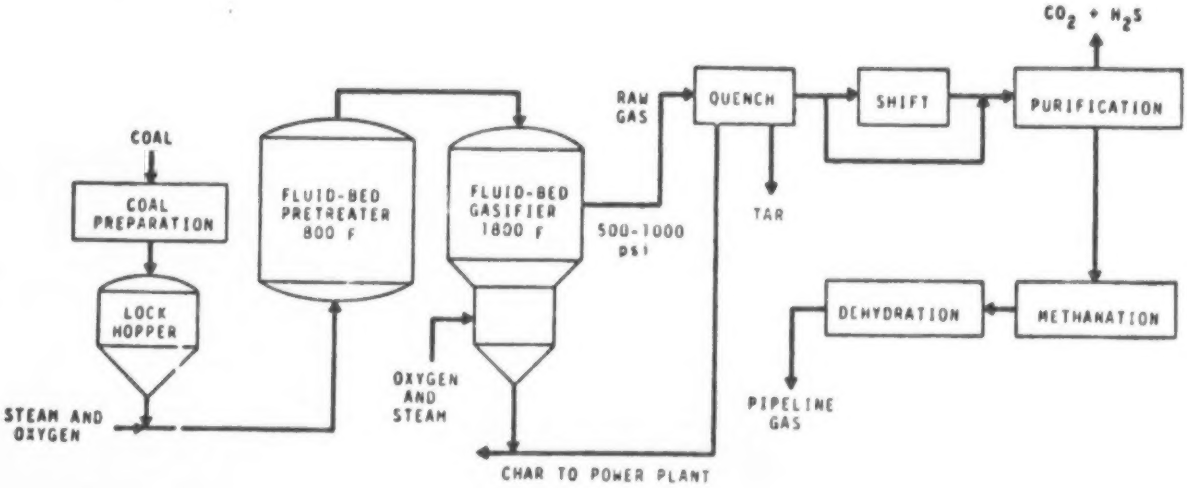

FIGURE 6.4. SYNTHANE PROCESS (4)

TABLE 6.4 THE SYNTHANE PROCESS (U.S. BUREAU OF MINES)

Facilty	Location	Owner(s) or Contractor	Status/Operating History
75 ton/day Pilot Plant (fully integrated to produce SNG)	Bruceton, Pa.	U.S. Bureau of Mines	Construction is about 60% completed as of July 1974. Expect completion in Jan. 1975. Cost $\$13 \times 10^6$. Studies on a wide variety of coals with a 40 lb/hr gasifier indicate process will accept any U.S. coal. Also, studying methanation using both parallel plate- and tube wall-type reactors with Raney nickel as the catalyst. Shift catalysts are also being studied.

(Contact: **(A. J. Forney, U.S. Bureau of Mines, Pittsburgh, Pa., (412) 892-2400)**

References:

(1) Hottel, H. C. and Howard, J. B., New Energy Technology - Some Facts and Assessments, MIT Press, Cambridge Mass (1971)

(2) "Evaluation of Coal Gasification Technology, Part I, Pipeline Quality Gas", R&D Report No. 74, Interim Report No. 1, Office of Coal Research, Washington, D.C. (1973).

(3) The Supply-Technical Advisory Task-Force-Synthetic Gas-Coal, prepared by Synthetic Gas-Coal Task Force for the Federal Power Commission (April, 1973).

(4) Forney, A. J., Haynes, W. P., Elliot, J. J., Gasior, S. J., Johnson, G. E. and Starkey, J. D., Jr., "The Synthane Coal-to-Gas Process". Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 199-208, Chicago (1973).

6.5 The Bi-Gas Process (Bituminous Coal Research)

Description of the Process. The primary gasification process is described in Section 5.5.1. After the gas from that process (operating with oxygen) is shifted to the proper hydrogen/carbon monoxide ratio, the hydrogen sulfide and carbon dioxide are selectively removed in a SELEXOL unit prior to catalytic methanation (Figure 6.5).

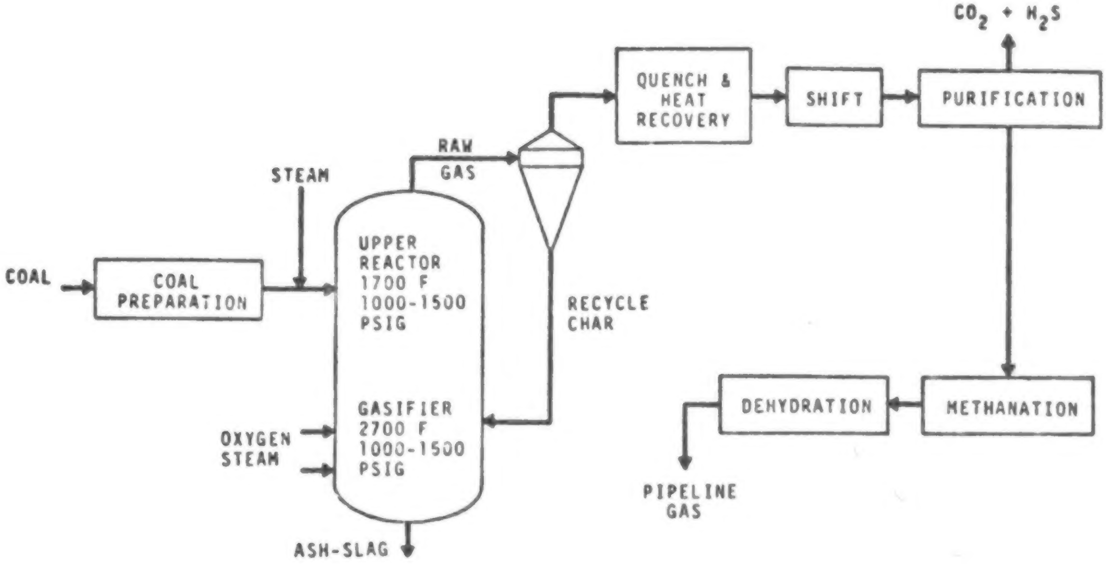

FIGURE 6.5. BIGAS PROCESS(4)

TABLE 6.5. BIGAS: STATE OF THE ART

Facility	Location	Owner(s) or Contractor	Status/Operating History
5 ton/hr Pilot	Homer City, Pa.	Bituminous Coal Research (with support from OCR and AGA). (Stearns Roger Corp. is responsible for construction).	Under construction. Estimate completion in early 1975. Estimated cost is about $\$25 \times 10^6$. This will be a fully integrated plant. Lab- and process development scale (6000 SCF/hr) studies of methanation are continuing to develop optimum operation conditions for the pilot plant. Acid gas removal by Selexol process.

(Contact: Robert Grace, Bituminous Coal Research, Pittsburgh, Pa. (412) 327-1600)

References:

(1) Hottel, H.C. and Howard, J.B., New Energy Technology: Some Facts and Assessments, MIT Press, Cambridge Mass. (1971).

(2) "Evaluation of Coal Gasification Technology. Part I, Pipeline Quality Gas", National Research Council, National Academy of Engineering, Washington, D.C. (1973).

(3) The Supply-Technical Advisory Task Force-Synthetic Gas-Coal, prepared by Synthetic Gas Coal Task Force for the Federal Power Commission (April, 1973).

(4) Grace, Robert J., "Development of the Bigas Process", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 179-198, Chicago, (1973).

(5) "Clean Energy from Coal--a National Priority", 1973 Annual Report, Office of Coal Research, Washington, D.C. (1973).

(6) "Coal Technology: Key to Clean Energy", Annual Report 1973-74, Office of Coal Research, Washington, D.C. (1974).

(7) Hegarty, W.P. and B.E. Moody, "Evaluating the Bigas SNG Process", Chemical Engineering Progress, Vol. 69, No. 3, pp 37-42 (1973).

6.6 The CO_2 Acceptor Process (Consolidation Coal Company)

Description of the Process. The primary gasification process is described in Section 5.3.3. Raw gas from the gasifier is purified and catalytically methanated (Figure 6.6). Because the purified gas has a high hydrogen/carbon monoxide ratio, shift conversion may not be necessary.

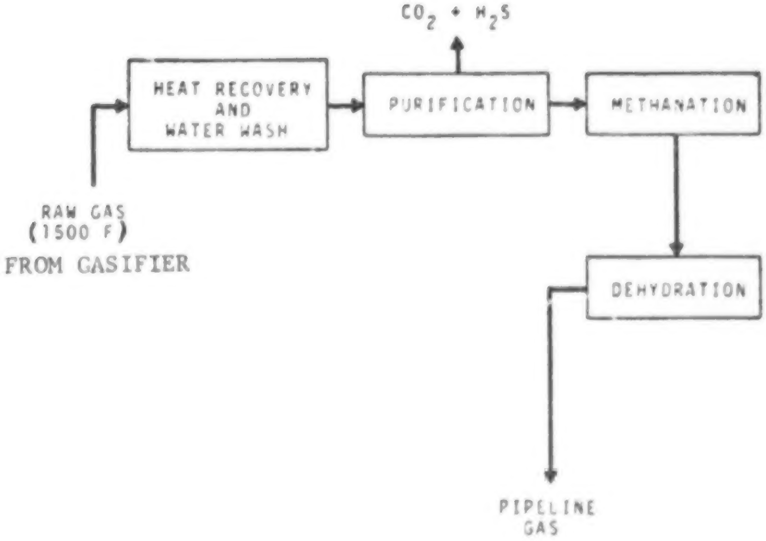

FIGURE 6.6. THE CO_2 ACCEPTOR PROCESS

TABLE 6.6. CO_2 ACCEPTOR: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
40 Ton/day Pilot plant	Rapid City, S.D.	Consolidation Coal Co., (OCR and AGA support) (plant constructed and operated by Stearns Roger Corp.)	- Plant cost about $\$9.3 \times 10^6$. In operation since April 1972, - 25 runs were completed in the period 4/72-5/74. Three continuous runs of 100 or more hours. - Mechanical problems in earlier runs have slowed the acquisition of fundamental data. More recent runs have been more successful. - Problem of refractory failures has been solved. - Corrosion of fired heaters by H_2S has been eliminated by use of ZnO system to remove H_2S. However, formation of metal carbides by carbon deposition still resulted in loss of metal. Hope to solve this by adding steam to the gas going to these burners. - Problems with char combustion and plugging of acceptor lines during start-up also appear to be solved. - Designed for use with lignite and subbituminous coal. - Designed to operate at pressure of 150 to 300 psi and temperatures up to 1800 F.
	(Contact: Carl E. Fink, Consolidation Coal Co., Rapid City, S.D., (605) 342-6416)		

References:

(1) Hottel, H. C., and Howard, J. B., New Energy Technology - Some Facts and Assessments, MIT Press, Cambridge, Mass., (1971).

(2) "Evaluation of Coal [illegible]fication Technology, Part I, Pipeline-Quality Gas, Office of Coal Research, Washington, D.C. [illegible]

(3) The Supply-Technica[illegible]isory Task Force-Synthetic Gas-Coal, prepared by Synthetic Gas-Coal Task Force for the Federal Power Commission (April, 1973).

(4) Fink, Carl E., "The CO_2 Acceptor Process", Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 301-310, Chicago (1973).

(5) Annual Report for Calendar Year 1972, Office of Coal Research, U.S. Dept. of Interior, Washington (1973).

(6) Annual Report 1973-74, Office of Coal Research, U.S. Dept. of Interior, Washington (1974).

6.7 The Hydrane Process

Description of the Process. This process (Figure 6.7) involves direct hydrogasification of raw coal in a rather unique two-stage hydrogasifier. Pulverized coal is fed into the upper stage which is a free-fall dilute-phase reactor operated at 1650 F and 1000 psi. The coal is devolatilized while flowing in dilute-phase suspension concurrently downward with a hot stream of gas which is about 50 percent hydrogen and 50 percent methane. Char from this stage flows into the second stage (fluidized-bed) where partial gasification and methanation occur in the presence of almost pure hydrogen to produce the feed gas for the upper stage. The result is an off gas rich in methane (about 70 volume percent) which requires only light methanation, and which is relatively simple to purify because of its low carbon dioxide content ($<1\%$). Hydrogen sulfide removed from the gas stream can be fed directly to a Claus plant without further reduction in carbon dioxide.

Residual char from the second stage is drawn off through a standpipe and is fed directly to the hydrogen-rich synthesis gas generator. Residual char from the hydrogen plant (0.137 lb/lb dry coal) may be used as fuel for steam and power generation since it contains enough carbon to be combustible.

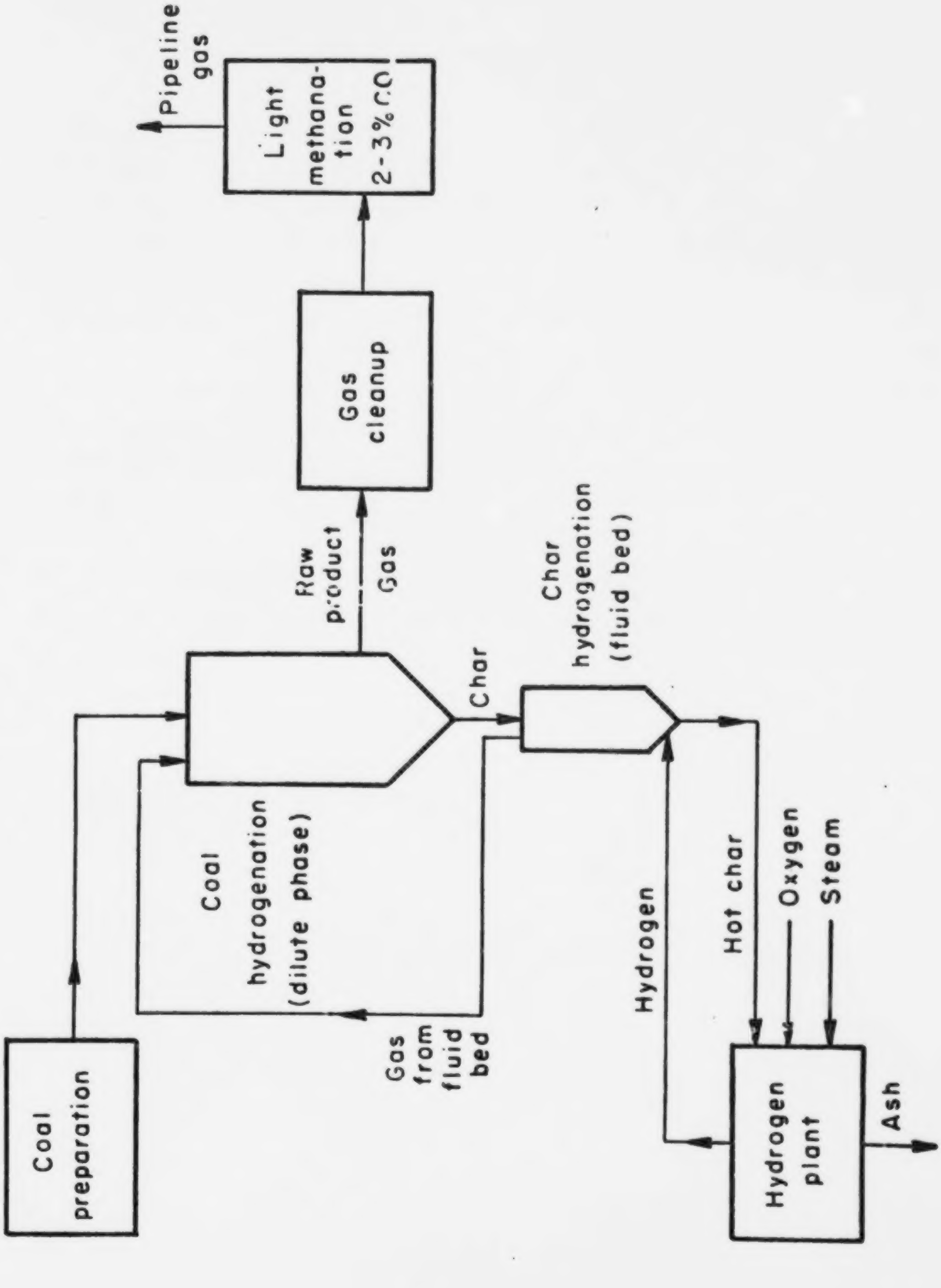

FIGURE 6.7. HYDRANE PROCESS(4)

TABLE 6.7 HYDRANE: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
1 Ton/hour Pilot	N.A.	U.S. Bureau of Mines	Under design. Free-fall dilute-phase reactor has been operated as a seperate lab-scale unit at pressures of 35 to 205 atm. and at temperatures up to 1650 F. Carbon conversion was satisfactory and the kinetics have been well-defined (3,6). Second-stage reactor also has been operated as a seperate unit on a bench scale. In past two years major emphasis has been operation of an integrated two-stage hydrogasifier (10lb/hr). Because of the equipment scale, early attempts at operating 2nd stage as a fluid bed were plagued with basic design problems. Nonetheless, steady state was attained for short periods in several runs. Total C conversion was over 50 percent for both stages. H_2 consumption per unit of CH_4 produced was low (1.38). Hope to develop a smooth-running unit to generate basic data on 2nd stage using freshly produced char from stage 1, to measure liquid product yields, and to prove operability. Also, obtaining data on production of hydrogen-rich synthesis gas from Hydrane char.

(Contact: James A. Gray, U.S. Bureau of Mines, Pittsburgh, Pa (412) 892-2400)

References:

(1) Hottel, H. C., and Howard, J. B., New Energy Technology, Some Facts and Assessments, Cambridge, Mass., MIT Press (1971).
(2) "Evaluation of Coal Gasification Technology. Part 1. Pipeline-Quality Gas", R&D Report No. 74 - Interim Report No. 1, Office of Coal Research, Washington, D.C. (1973).
(3) Feldman, H. F., Mima, J. A., and Yavorsky, P. M., "Pressurized Hydrogasification of Raw Coal in a Dilute-Phase Reactor", 165th Annual ACS Meeting, Dallas, Texas, April 1973. (Preprinted).
(4) Yavorsky, Paul M., "The Hydrane Process", p 209, Institute of Gas Technology Symposium on Clean Fuels from Coal, Chicago (1973).
(5) Feldmann, J. F., Wen, C. Y., Simons, W. H., Yavorsky, P. M., "Supplemental Pipeline Quality Gas from Coal by the Hydrane Process", Paper presented at the 71st National Meeting AICE, Dallas (February 20-23, 1972).
(6) Feldmann, H.F., Simons, W. H., Mima, J. A., and Hiteshue, R. W., Preprints of Fuel Div., ACS, Chicago (Sept. 1970).

6.8 The Ash Agglomeration Process (Union Carbide-Battelle)

Description of the Process. The primary gasification process is described in Section 5.4.3. Raw gas from this fluidized bed gasifier passes through a heat recovery system and is expanded in a gas turbine to generate process power. This gas is then shifted, purified and catalyticly methanated (Figure 6.8).

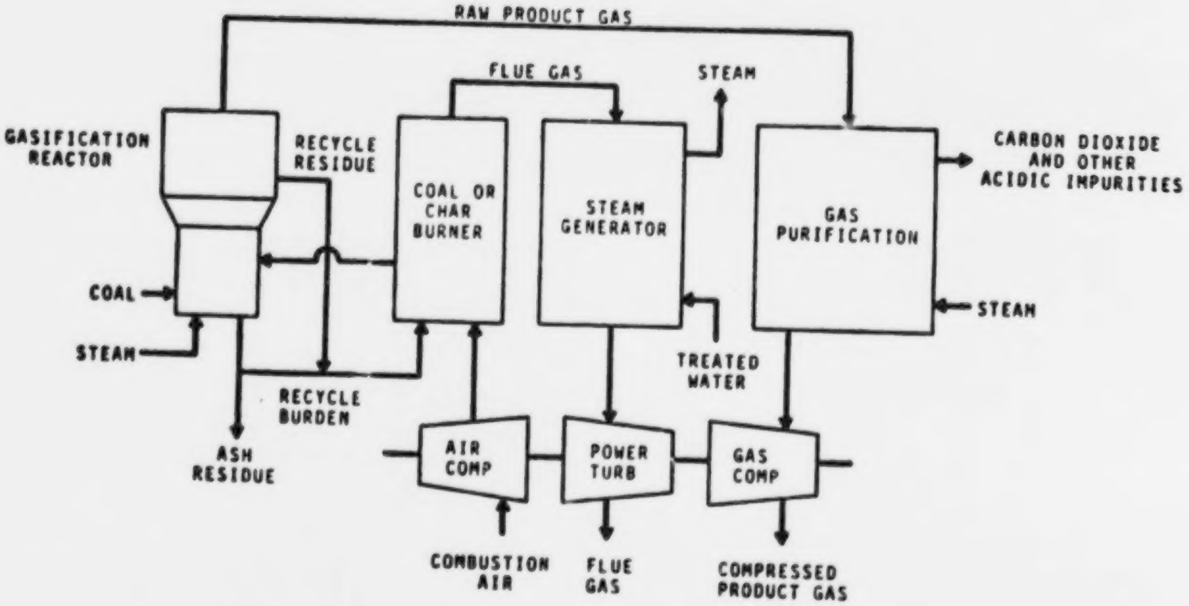

FIGURE 6.8. UNION CARBIDES' AGGLOMERATED ASH PROCESS(3)

TABLE 6.8. ASH AGGLOMERATION

Facilty	Location	Owner(s) or Contractor	Status/Operating History
1200 lb/hr Process Development Unit	West Jefferson, O	Battelle (Funded by OCR and AGA) (Patent for the process is held by Union Carbide.)	Under construction. Estimate completion in first quarter of 1975. Earlier studies included bench-scale studies of both the coal burner and the gasifier. Erosion by fly ash from the burner at simulated turbine conditions was also investigated. The Process Development Unit will not include systems for gas purification, shift conversion or methanation.

(Contact: W. M. Goldberger, Battelle Columbus Labs., (614) 299-3151)

References:

(1) Corder, W. C., Batchelder, H. R., and W. M. Goldberger, "The Union Carbide/Battelle Coal Gasification Process Development Unit Design", Presented at the Fifth Synthetic Pipeline Gas Symposium, Chicago (Oct. 1973).

(2) "Evaluation of Coal-Gasification Technology, Part II. Low- and Intermediate- Btu Fuel Gas", Report by National Research Council, National Academy of Engineering (1973).

(3) Goodridge, E., "Status Report: The AGA/OCR Coal Gasification Program", Coal Age 78, 54-59 (January 1973).

6.9 The Kellogg Molten Salt Process (M. W. Kellogg Co.)

Description of the Process. The primary gasifier system is described in Section 5.7.2. After the gas leaving the gasifier is processed to recover heat and entrained salt, it is passed through a shift converter to increase the ratio of hydrogen to carbon monoxide. It is subsequently purified and catalytically methanated.

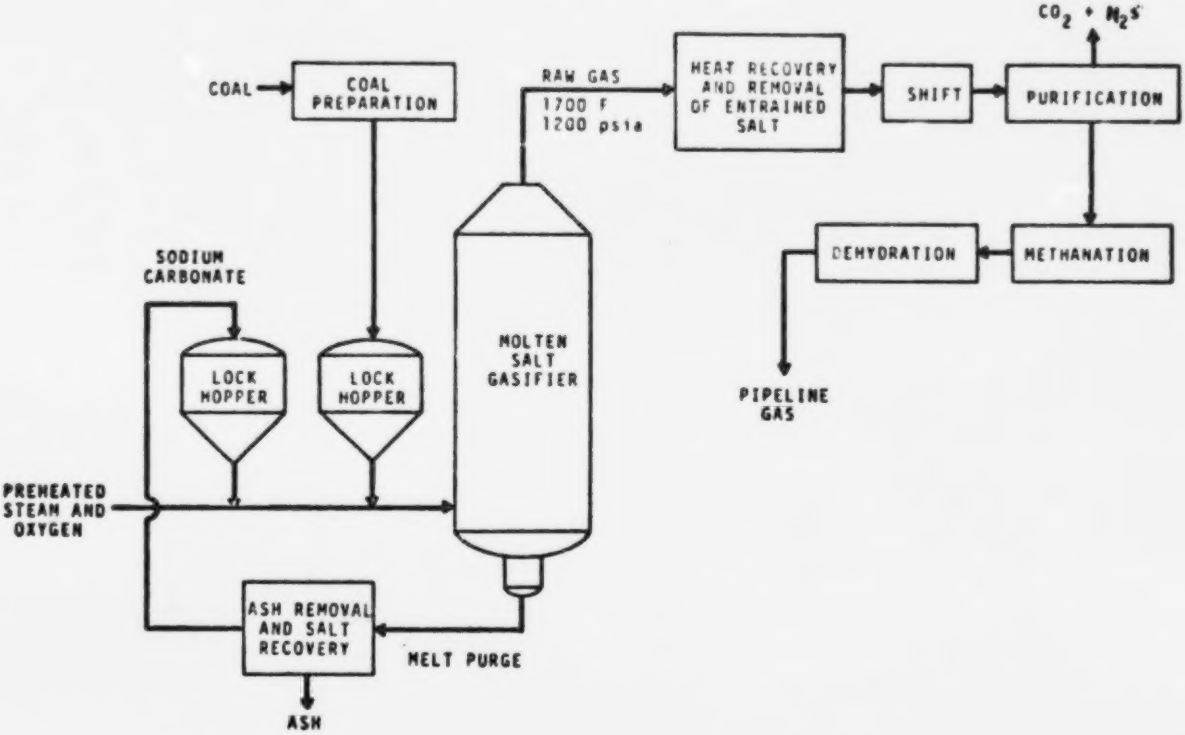

FIGURE 6.9. KELLOGG MOLTEN SALT PROCESS(2)

TABLE 6.9. KELLOGG MOLTON SALT: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
-------	------	M.W. Kellogg Company	Bench scale studies only. The molten salt creates serious corrosion problems, but these have been solved by use of an aluminum oxide refractory, Monofrax A. Report no corrosion after 600-700 hrs continuous operation. A process development unit is planned. Preliminary flow sheets and cost estimates have been made. Seeking support.

(Contact: A. E. Cover, M. W. Kellogg Company, Houston, Texas, (713) 626-5600).

References:

(1) Cover, A. E. and Schreiner, W. C., The Kellogg Molten Salt Process, Presented at the 4th Conference on Synthetic Fuels from Coal, Oklahoma State University, Stillwater, Oklahoma (1974).

(2) Cover, A. E., Schreiner, W. C., and Skaperdas, G. T., The Kellogg Coal Gasification Process: Single Vessel Operation, Institute of Gas Technology Symposium on Clean Fuels from Coal, pp 273-284, Chicago (1973).

(3) Evaluation of Coal-Gasification Technology, Part I, Pipeline-Quality Gas, National Research Council, National Academy of Sciences, Washington, D.C. (1973).

6.10 Atgas Process (Applied Technology Corp.)

The primary gasification is essentially the same as that described earlier in Section 5.7.1 for production of intermediate-Btu gas in a molten iron bath. The intermediate Btu gas is upgraded by the water-gas shift reaction followed by purification and catalytic methanation.

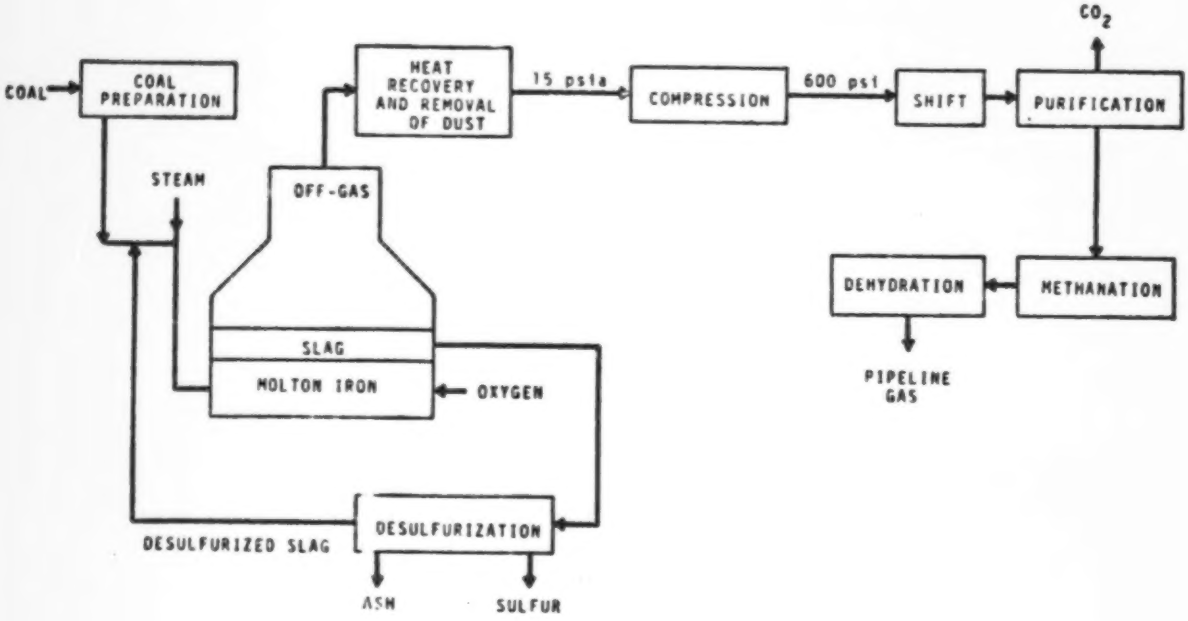

FIGURE 6.10. ATGAS PROCESS(1)

TABLE 6.10. MOLTEN IRON: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
-------	-------	Applied Technology Corp.	Pre-pilot stage. Studies have involved a 25 inch I.D. induction furnace (4000 lb capacity) to simulate the gasifier, and using air to produce low-Btu fuel gas. The off gas handling system was equipped to permit continuous analysis for SO_2, NO_2, NO, H_2, O_2, CO_2 and CO. Expendable, non-cooled ceramic lances were used for injection. Results indicate a boiler feed gas with less than 50 ppm SO_2 can be generated from high-S coal (3.5 percent)(1)

(Contact: Ronald J. McGarvey, Applied Technology Corp., Pittsburgh, Pa (412) 782-0682)

References:

(1) LaRosa, Paul and McGarvey, Ronald J., "Fuel Gas from Molten Iron Coal Gasification", Institute of Gas Technology Symposium on Clean Fuels, pp 285-3000, Chicago (1973).

(2) "Evaluation of Coal Gasification Technology, Part I, Pipeline-Quality Gas, National Research Council, National Academy of Engineers, Washington, D.C. (1973).

(3) "Clean Energy from Coal: A National Priority", 1973 Annual Report (for Calendar Year 1972) Office of Coal Research, Washington, D.C. (1973).

6.11 Garrett Flash Pyrolysis (Garrett Research and Development)

Description of the Process. The primary gasification process is described in Section 5.5.4. The raw pyrolysis gas (600 to 650 Btu/SCF) is separated from char in a series of cyclones and is upgraded to pipeline quality by shift conversion, purification and catalytic methanation. The process produces about 8,500 SCF/ton of coal (for pyrolysis at 1700 F).

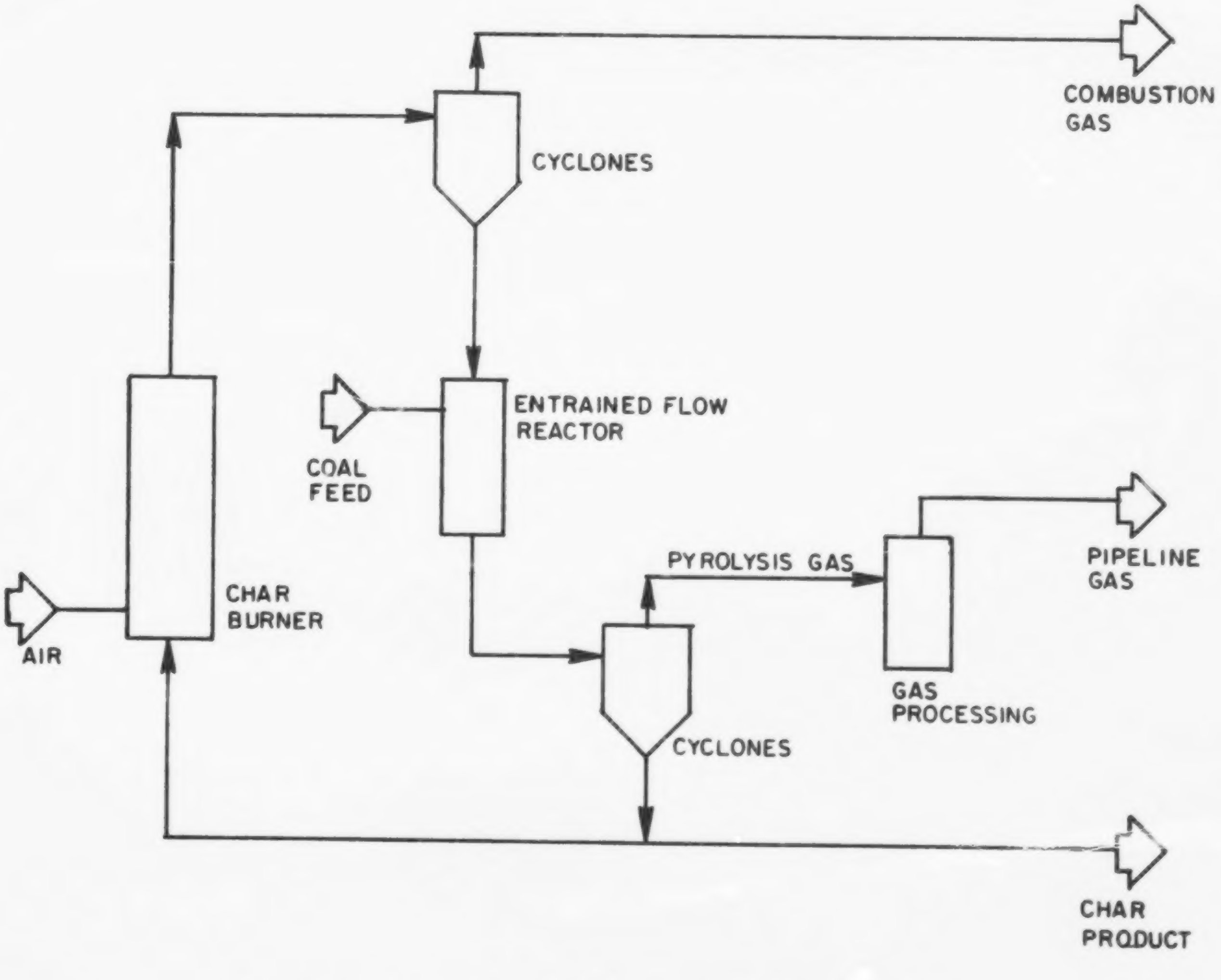

FIGURE 6.11. GARRETT FLASH PYROLYSIS PROCESS(2)

TABLE 6.11. GARRETT FLASH PYROLYSIS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
10 Ton/hr Pilot	LaVerne, Ca.	Garrett Research and Development (research subsidiary of Occidental Petroleum)	Proposed. Seeking support. A bench scale pyrolysis unit (50 lb/hr) has been in operation since Jan. 1973 (2).

(Contact: D. E. [illegible]as, Garrett Research and Development, La Verne, Ca., (714) 593-7421)

References:

(1) McMath, H. G., Lumkin, R. E., and Sass, A., "Production of Gas from Western Sub-Bituminous Coals by the Garret Flash Process", Presented at the 66th Annual Meeting AICHE Philadelphia, Pa. (November 1973).

(2) McMath, H. G., Lumkin, R. E., Longanbach, J. R., and Sass, A., "A Pyrolysis Reactor for Coal Gasification", Chemical Engineering Progress, Vol. 70, No. 6, pp 72-3 (June 1974).

(3) Adams, D. E., Sack S., and Sass, A., "Coal Gasification by Pyrolysis", ibid pp 74-75.

(4) Adams, D. E., Sack S., and Sass, A., "The Garret Pyrolysis Process", presented at the 66th Annual Meeting AICHE, Philadelphia, Pa. (November 15, 1973).

6.12 Cogas (Cogas Development Company)

Description of the Process. A brief description of the gasification process based on the limited information available is given in Section 5.3.6.

The raw gas from the gasifier is converted to SNG by shift-conversion, purification and catalytic methanation. Since the gasification is carried out at low pressures, the gas must be compressed to meet pipeline specification.

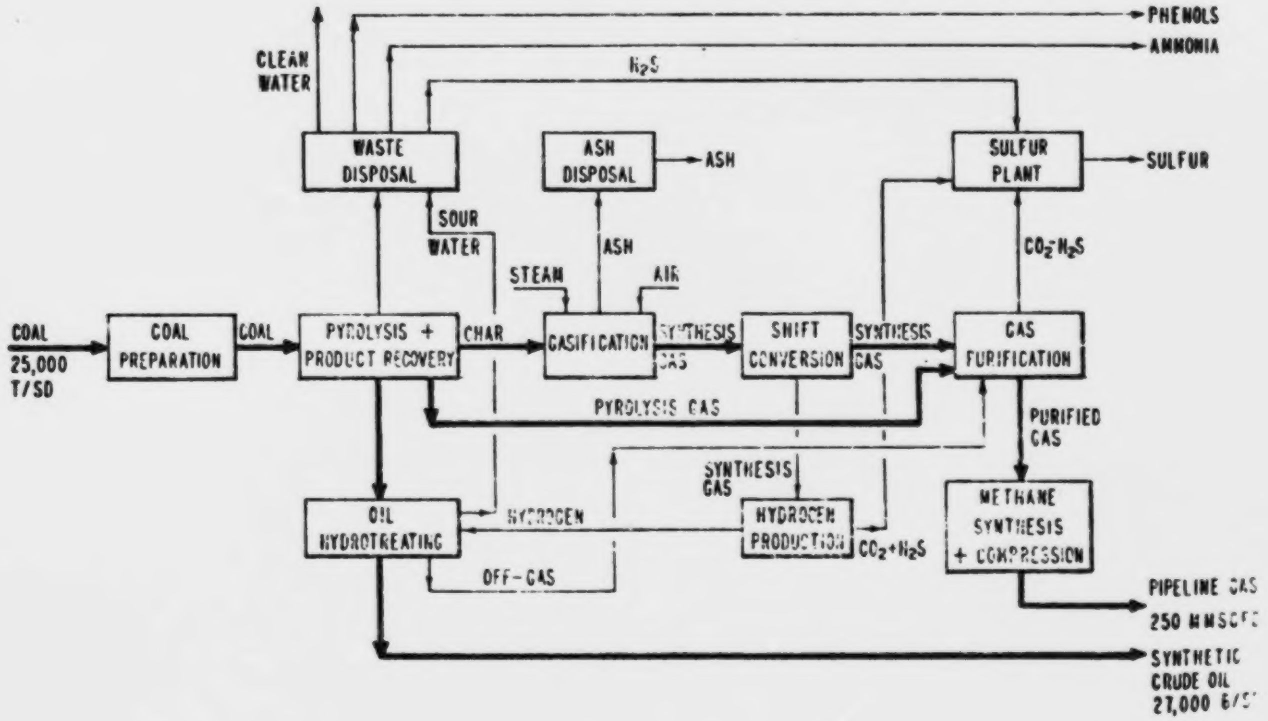

FIGURE 6.12. A COGAS PROCESS DIAGRAM

TABLE 6.12. COGAS: STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
100 Ton/day Pilot	Leatherhead, England	Operated for Cogas Development Co. by the British Coal Utilization Board	Operational since March 1974. Mechanical problems were few and were overcome with relative ease. Past several months have been devoted to proving process operability.
5 Ton/day Pilot	Plainsboro, N.J.	Cogas Development Co. (a consortium of FMC Corp., Consolidated Natural Gas, Panhandle Eastern Pipeline, Republic Steel, Rocky Mountain Energy and Tenn. Gas Transmission Co.)	Operational since late spring of 1974. No information available.

(Contact: Howard Melakoff (General Manager), Cogas Development Co., Princeton, N. J. (609) 452-2300

References:
(1) Dierdorff, L. H., Jr., and Bloom, R., Jr., "The COGAS Project - One Method of Coal to Gas Conversion", paper presented at the West Coast Meeting of the Society of Automotive Engineers, Portland, Oregon (August 20-23, 1973).
(2) Perry, Harry, "Coal Conversion Technology", Chemical Engineering, pp 88-102 (July 22, 1974).

Supplemental Bibliography for Coal Gasification(1)

(1) Hottel, H. C. and Howard, J. B., New Energy Technology, Some Facts and Assessments, Cambridge, Mass., MIT Press (1971).

(2) Von Fredersdorff, C. G., Elliot, M. A., "Coal Gasification" in Chemistry of Coal Utilization, H. H. Lowey, Editor, Supplementary Vol., John Wiley and Sons, Inc., New York, pp 892-1022 (1963).

(3) The Supply-Technical Advisory Task Force - Synthetic Gas-Coal, prepared by Synthetic Gas-Coal Task Force for the Federal Power Commission (April 1973).

(4) Bituminous Coal Research, Inc., "Gas Generator Research and Development Survey and Evaluation-Phase One", R&D Report No. 20, Interim Report No. 1, U.S. Dept. of Interior, Office of Coal Research, Washington, D.C. (August 1965).

(5) Waterman, W. W., "Summary Presentation of an Overview of Coal Conversion Technology", IGT Symposium on Clean Fuels from Coal, pp 673-682, Chicago (1973).

(6) "Report to Project Independence Blueprint, Federal Energy Agency", Prepared by the Interagency Synthetic Fuels Task Force, Supplement 1 (July 8, 1974).

(7) "Evaluation of Coal Gasification Technology. Part I. Pipeline Quality Gas", National Research Council, National Academy of Engineering, Washington, D.C. (1973).

(8) "Evaluation of Coal Gasification Technologies, Part II, Low- and Intermediate-Btu Fuel Gases", National Research Council, National Academy of Engineering, Washington, D.C. (1973).

(9) Bodle, W. W., Vyas, K. C., "Clean Fuels from Coal-Introduction to Modern Processes", IGT Symposium on Clean Fuels from Coal, pp 49-91 Chicago (1973).

(10) Wen, C. Y., Editor, "Optimization of Coal Gasification Processes", R&D Report No. 66, Interim Report No. 1, U.S. Dept. of Interior, Office of Coal Research, Washington, D.C. (1972).

(11) Connor, Jack G., "Coal Gasification: A Review of Status and Technology", paper presented to AAAS National Meeting, San Francisco (Feb. 26, 1974).

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(1) References to specific processes appear in the individual sections of Sections 5 and 6.

(12) Wen, C. Y., Li, C. T., Tscheng, S. H. and O'Brien, W. S., "Comparison of Alternative Coal-Gasification Processes for Pipeline Gas Production", Energy Sources, 1(1), 31 (1973).

(13) Perry, Harry, "Coal Conversion Technology", Chemical Engineering, pp 88-102 (July 22, 1974).

(14) Chopey, Nicholas P., "Gas-from-Coal: An Update", Chemical Engineering, pp 70-73 (March 4, 1974).

(15) Anon, "New Processes Brighten Prospects of Synthetic Fuels from Coal", Engineering and Mining Journal, Vol 75, No. 175, pp 91-97 (April 1974).

(16) Hale, Dean, "Coal Gasification Takes on a New Look", Pipeline and Gas Journal, pp 23-26 (March 1974).

(17) "Coal Gasification--The Future Fuel Source", Gas Turbine International, pp 19-24 (May-June 1974). (Excerpted from Office of Coal Research 1973-1974 Annual Report.)

(18) Levene, Harold D., "Gasification or Liquifaction: Where We Stand", Coal Mining and Processing, pp 43-48 (January 1974).

(19) Goodridge, Edward, "Status Report: The AGA/OCR Coal Gasification Programs", Coal Age, pp 54-59 (January 1973).

(20) Qader, S. A., "Low-Btu Gas Production from Coal", Intermet Bulletin, 4(3), 36-42 (1974).

(21) Quader, S. A., "SNG Production", Intermet Bulletin, 2(3), 34-38 (1973).

(22) Huebler, Jack, "Coal Gasification: State of the Art", Heating/Piping/Air Conditioning, 45(1), 149-155 (1973).

(23) Seigel, H. M. and Kalina, T., "Coal-Gasification Costs May lower", The Oil and Gas Journal, pp 87-94 (February 12, 1973).

(24) Mehta, D. C. and Crynes, B. L., "How Coal-Gasification Commonbase Costs Compare", The Oil and Gas Journal, pp 68-71 (February 5, 1973).

(25) Agosta, J., et. al., "Status of Low-Btu Gas as a Stragety for Power Station Emission Control", AICHE 65th Annual Meeting, New York (November 1972).

(26) Agosta, J., et.al., "The Future of Low-Btu Gas in Power Generation", Proceedings of the American Power Conference, 35 510-22 (1973).

(27) Ball, D., Smithson, G., Engdahl, R., and Putnam, A., "Study of Potential Problems and Optimum Opportunities in Retrofitting Industrial Processes to Low- and Intermediate- Energy Gas from Coal", Prepared by Battelle-Columbus Laboratories for Office of Research and Development, U.S. EPA, Washington, D.C. (May 1974).

(28) Magee, E. M., Jahnig, C. E., and Shaw, H., "Evaluation of Pollution Control in Fossil Fuel Conversion Processes", Prepared by Esso Research and Engineering Company for Office of Research and Development, U.S. Environmental Protection Agency, Washington, D.C. (January 1974).

(29) Ashworth, R. A., and Hsieh, B. C., "Low Btu Gasification of Coal: Who Needs it and How Can It be Improved?", presented at the EPA Symposium on Environmental Aspects of Fuel Conversion Technology, St. Louis (May 13-16, 1974).

(30) Rubin, E. S. and MCMichael, "Some Implications of Environmental Regulating Activities on Coal Conversion Processes", ibid.

(31) "Environemntal Considerations in Future Energy Growth", Volume 1, Report prepared by Battelle Memorial Institute for the Office of Research and Development, EPA (April 1973).

(32) Forney, A. J., et.al., "Analyses of Tars, Chars, Gases and Water Found in Effluents from the Synthane Process", U.S. Bureau of Mines, Technical Progress Report 76 (January 1974).

(33) "U.S. Energy Outlook: Coal Availability", A Report by the Coal Task Group, National Petroleum Council (1973).

(34) "Project Independence: An Economic Evaluation", MIT Energy Laboratory, Printed in Technology Review, pp 26-58 (May 1974).

(35) Siegel, Howard M., "The Cost and Commercialization of Gas and Liquids from Coal", IGT Symposium on Clean Fuels from Coal, pp 653-661, Chicago (1973).

(36) Siegel, H. M. and Kalina, T., "Technology and Cost of Coal Gasification", Mechanical Engineering, pp 23-28 (May 1973).

(37) Moe, J. M., "SNG From Coal via the Lurgi Process", IGT Symposium on Clean Fuels from Coal, pp 91-105, Chicago (1973).

(38) "Development of Information for Standards of Performance for the Fossil Fuel Conversion Industry", prepared for Industrial Studies Branch, EPA, by Battelle, Columbus Laboratories (June 21, 1974).

(39) Anon, "Expert Examines Materials Problems", Chemical Engineering, pp 58-59 (July 22, 1974).

(40) "Gas Industry Research Plan: 1974-2000", American Gas Association (January 1974).

(41) Robson, F. L., "Fuel Gasification and Advanced Power Cycles- A Route to Clean Power", presented at the Third International Conference on Fluidized Bed Combustion, Vol. 11, pp 205-225 (December 1973).

(42) Matthews, C. W., "A Design Basis for Utility Gas from Coal", ibid, pp 229-245.

(43) Crouch, W. B., et al., "Recent Experimental Results on Gasification Combustion of Low-Btu Gas for Gas Turbines", Combustion, pp 38 (April 1974).

(44) A. D. Little, Inc., "A Current Appraisal of Underground Coal Gasification", U.S. Dept. of the Interior, Bureau of Mines, Washington, D.C. PB 209 274, NTIS, Springfield, Va., 280 p (April 1972).

(45) Nadkarni, R. M., Bliss, C. and Watson, W. I., "Underground Gasification of Coal", Institute of Gas Technology Symposium on Clean Fuels from Coal, Chicago, pp 611-637 (1973).

(46) Schrider, L. A., and Pasini, J., Underground Gasification of Coal -- Pilot Test, Hanna, Wyoming, presented at the AGA Fifth Synthetic Pipeline Gas Symposium (October 1973).

(47) Linden, Henry R., "The Role of SNG in the U.S. Energy Balance", presented at the IGT Symposium on SNG from Hydrocarbon Liquids, pp 55-76 (March 1973).

SECTION 7

FLUIDIZED BED COMBUSTION

7.1 Atmospheric Fluidized-Bed Combustion

Description of the Process. The combustion of solid fuels takes place in a fluidized-bed of limestone (or dolomite), solid fuel and ash at 2-15 ft/sec and temperatures of 1400-1800 F. About 50 percent of the combustion heat can be removed by the immersed heat transfer tubes and the surrounding water walls. The high heat transfer rate in the fluidized bed (50 Btu/hr ft^2 F as compared to 10-15 Btu/hr ft^2 F in conventional boilers) resulted in a high volumetric heat release rate of 5,000,000 Btu/hr ft^3 in the combustion space as compared to 20,000 Btu/hr ft^3 in a pulverized-coal-fired boiler. This feature makes the fluidized bed boiler compact enough to allow complete or partial shop fabrication of the boiler, thus reducing significantly the capital investment.

Since the fluidized-bed boiler is operated at 1400-1800 F, the SO_2 released during the combustion by the high sulfur fuel can be ideally absorbed by the limestone or dolomite present in the bed. To reduce the solid waste disposal burden, the spent limestone may be regenerated at slightly reducing conditions at high temperatures (2000 F). To increase the overall combustion efficiency, the elutriated carbon would be combusted in a separate combustor at a much higher temperature (2000 F) (Carbon Burn-up Cell). The bench and pilot data indicate that NO_x emission is low and on the order of 260 ppm. Over 90 percent SO_2 removal and an overall combustion efficiency of over 98 percent can be achieved.

The conceptual design of modular boiler concepts of single bed level and packed bed configuration has been studied for utility and industrial applications. A schematic diagram of an atmospheric pressure fluidized bed combustion power plant is shown in Figure 7.1.

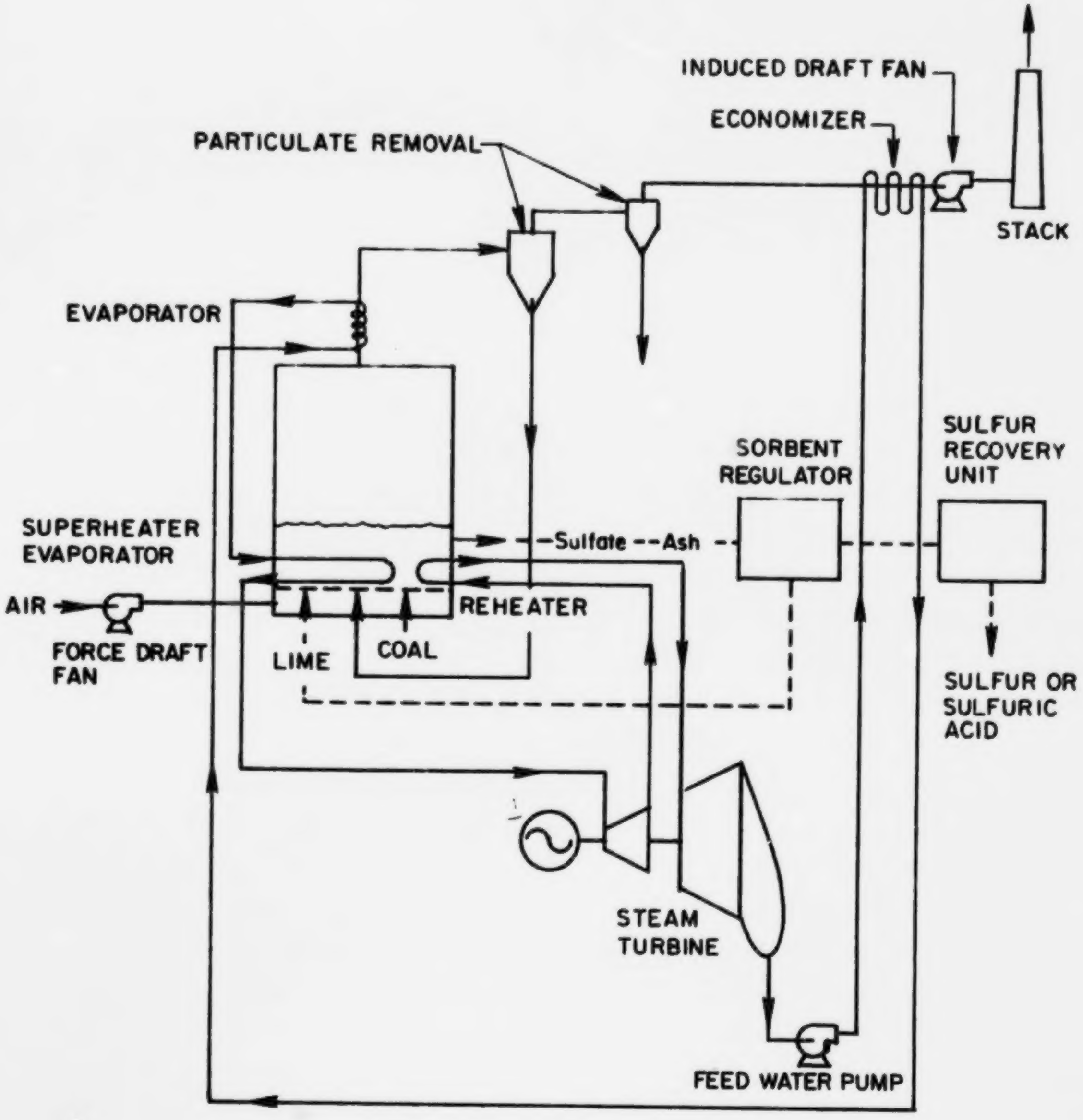


FIGURE 7.1. ATMOSPHERIC FLUIDIZED-BED COMPUSTION POWER PLANT

TABLE 7.1. ATMOSPHERIC FLUIDIZED-BED COMBUSTION STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
1. C.R.E. Pilot Combustor (3' x 3', 1 MW)	Cheltenham, U.K.	National Coal Board-Coal Research Establishment	- Bench scale work since 1963, pilot unit in operation since 1970, development work aimed at utility and large-scale boiler application (2fps), conceptual design studied by Babcock & Wilcox Ltd., Preece, Caradew, and Rider Ltd.
2. BCURA Pilot Combustor (27" dia)	Leatherhead, U.K.	BCURA Limited	- Research began in 1964, pilot unit in operation since 1966, development effort aimed at small industrial application (10 - 14 fps), conceptual design studied by John Thompson Ltd.
3. PER Pilot Combustor (12" x 16", 20" x 6')	Alexandria, Virginia	Pope, Evans & Robbins (Funded by OCR & EPA)	- Bench & pilot work since 1965, aimed at development of packaged coal burned boiler, the pollution control studies were supported by EPA - Pilot data have demonstrated 3-cell concept: boiler (1400 - 1800 F, 3% excess air), carbon burn-up cell (2000 F, >3% excess air), and regenerator (>1850 F, <.5% excess air). All operated at 6-14 fps
4. ANL Bench Combustor (6")	Chicago, Illinois	Argonne National Laboratories (EPA)	- In operation 68-72, fundamental combustion studies
5. B.M. Pilot Combustor (18")	Morgantown, West Virginia	Bureau of Mines (EPA-BM)	- In operation 68-72, studies on heat transfer, coal, and boiler tubes
6. Foster-Wheeler Cold Unit (6'x6')	Livingston, New Jersey	Foster-Wheeler Co. (OCR)	- Studies on coal feeding and tube bank design
7. 30 MW Multicell Fluidized-Bed Boiler	Rivesville, West Virginia	Pope, Evans & Robbins (OCR)	- Boiler dimension = 12' x 38' x 25' - Construction in progress since 1974, to be completed in 1976 - Plant cost is estimated at $\$11.0 \times 10^6$ with annual operating cost at $\$1.5 \times 10^6$ - To demonstrate the operability and reliability of a large fluidized-bed combustion system.

Contact: J. W. Bishop, Pope, Evans & Robbins, Inc. 320 King Street, Alexandria, Virginia 22314.

TABLE 7.1. (Continued)

References:

(1) N.C.B. Final report to EPA, "Reduction of Atmospheric Pollution", Vol. I, II, III, (1971).

(2) Skinner, D. G., "The Fluidized-Bed Combustion of Coal", Mills & Boon Limited, (1971).

(3) The 2nd and 3rd International Conference on Fluidized-Bed Combustion Syposium at Hueston Woods, Ohio (1968, 1970).

(4) Jonke, A. A., et al., ANL Report to EPA, ANL/ES-CEN-1004 (1971).

(5) Robison, E. B., et al., PER reports to OCR and EPA (1972, 1970).

(6) Annual Report 1973-74 of Coal Technology (Key to Clean Energy), OCR, U.S. Department of the Interior, Washington, D.C. (1974).

7.2. Pressurized Fluidized-Bed Combustion

Description of the Process. The combustion system is similar to atmospheric fluidized-bed combustion except that it operates at 10 atm pressure. The pressure operation reduces the boiler size significantly. To increase the heat transfer surface, deep bed combustion with immersed tube banks are proposed. High temperature compressed air is directed to a gas turbine to recover the heat energy. This operation also serves to reduce the heat transfer surface requirement.

In the pressurized combustion, a much lower NO_x emission (150 ppm) has been demonstrated. Over 99 percent combustion efficiency and sulfur removal has been established by BCURA experiments at low bed velocities (2 ft/sec) and with a Ca/S ratio of 1.6.

The prospect of reduced corrosion and erosion to the steam tubes at low fluidized combustion temperatures provides a potential of advanced steam condition. The gas turbine inlet temperature may also be increased. Therefore, a thermal efficiency up to 47 percent is feasible for a pressure fluidized-bed combustion combined cycle plant, which may be compared with 35 to 38 percent achieved by conventional steam-electric power plants.

The critical elements to the pressurized combustion at the present development stage is the demonstration of effective hot gas clean-up, thus, a reasonable turbine blade life may be expected, and the demonstration of deep bed combustion of immersed steam tubes.

A schematic diagram of a pressurized fluidized-bed combustion power plant is shown in Figure 7.2.

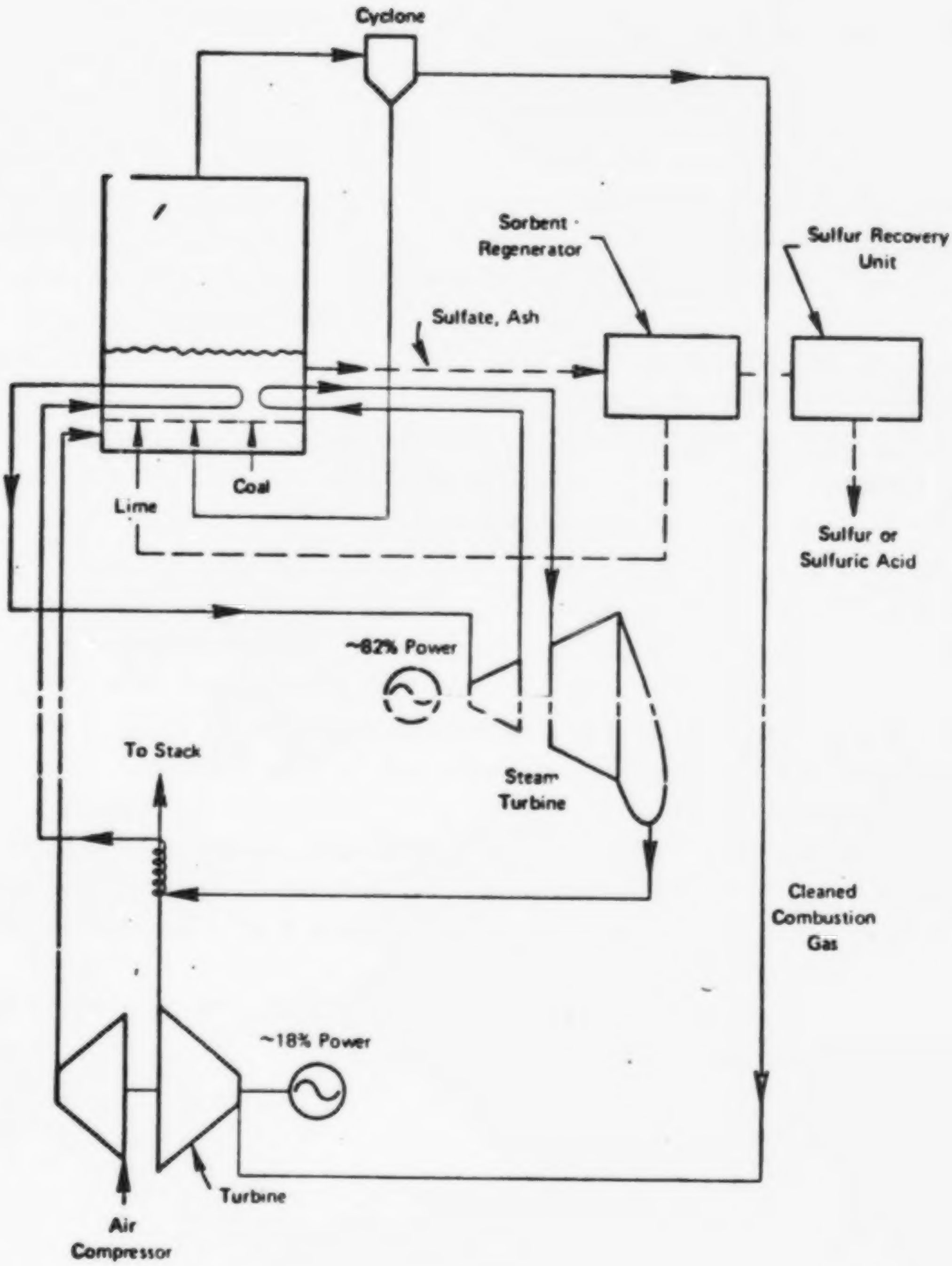


FIGURE 7.2. PRESSURIZED FLUIDIZED-BED COMBUSTION POWER PLANT

TABLE 7.2. PRESSURIZED FLUIDIZED-BED COMBUSTION STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
1. BCURA Pilot Combustor (48" x 24")	Leatherhead, U.K.	BCURA Limited	- In operation since 1969, designed at 6 atm - Excellent combustion efficiency and SO_2 and NO_x control have been demonstrated at low gas velocity (2 fps) and temperatures 1470-1750 F (Work jointly supported by NCB-EPA and NCB-OCR) - At low temperature (< 1600 F), there was no significant deposition on turbine blade cascade, but sufficiently extensive at bed temperature of 1750 that constant blade cleaning is required for continuous operation
2. ANL Bench Combustor (6" diam)	Chicago, Illinois	Argonne National Laboratories (EPA)	- Extensive fundamental data have been generated - Proposed one-stage and two-stage regeneration schemes were proved not feasible
3. ESSO Bench Combustor (3" diam)	Linden, New Jersey	EXXON Rd.E. (EPA)	- Combustion and regeneration study since 1968
4. ESSO Minplant (12.5" dia ~.65 MW)	Linden, New Jersey	EXXON Rd.E. (EPA)	- In operation at present without regeneration unit - Unit designed at 10 atm - Recent data appears to indicate that quality of fluidization needs to be improved - A turbine blade cascade is being designed by Westinghouse and to be added to the Miniplant - Deep bed combustion with immersed tube bank and effective hot gas clean-up needs to be demonstrated
5. CPU-400 Pilot Plant Unit (7' dia. 4atm, ~1.8 MW)	Menlo Park, California	Combustion Power Company	- Originally designed for refuse burning, without immersed tube bank, the modification has been made to burn coal (funded by OCR) - The granular filter for hot gas clean-up is to be installed for later refuse and coal tests.

Contacts: Hoke, R. EXXON Rd.E., Linden, New Jersey and Furlong, D. L., Combustion Power Company, Menlo Park, California.

TABLE 7.2. (Continued)

References:

(1) Skinner, D. G., "The Fluidized-Bed Combustion of Coal", Mills and Boon Limited (1971).

(2) Jonke, A. A., et al, "Reduction of Atmospheric Pollution by the Application of Fluidized-Bed Combustion", Annual Report (1974).

(3) BCURA, "Pressurized Fluidized-Bed Combustion", Report to OCR (1973).

(4) Molen, R. H. V., "Operational Experience with the CPU-400 Pilot Plant", Paper presented at 78th AIChE National Meeting, Salt Lake City, Utah (1974).

7.3. Ignifluid Combustion

Description of the Process. The Ignifluid process was invented by A. A. Godel in the late 40's and later developed by Societe Anonyme Activit and Babcock-Atlantique in the 1950's. A conventional stoker is modified to create a fluidized bed on the grate. The fluidized bed is operated between 1000 and 1200 C to agglomerate coal ash which is carried out of the bed on the moving grate. Partial combustion occurs in the bed and secondary air (~50 percent) is supplied above the bed to complete the combustion. The system offers low particulate emission as more than 50 percent of coal ash is removed as clinker from the grate. However, the Ignifluid process is deprived of the SO_2 and NO_x control potential, which is favored at much lower combustion temperatures.

There are four Ignifluid boiler plants in operation. A typical Ignifluid utility boiler is shown in Figure 7.3.

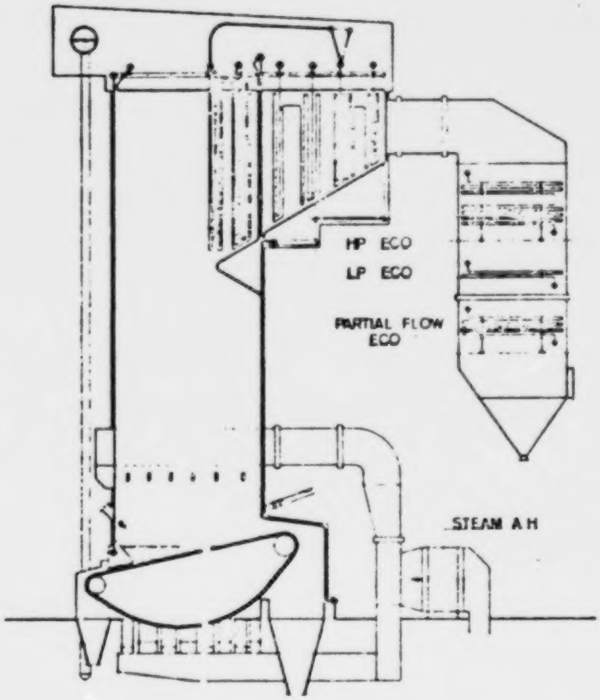


FIGURE 7.3. IGNIFLUID UTILITY BOILER

TABLE 7.3. IGNIFLUID COMBUSTION STATE OF THE ART

Facility	Location	Owner(s) or Contractor	Status/Operating History
Semi-industrial Ignifluid plant	Vernon, France	Societe Anonyme Activit/Babcock-Atlantique	- Combustion test began in 1953 - Commercial operation since early 1960's, 4 Ignifluid boiler plants in operation, and proposals nave been made to build a 250 MW power plant in Europe and large scales in the United States - SO_2 and NO_x control needs further development
Ignifluid Boiler Plants	La Taupe	Babcock-Atlantique	- 77,000 lb/hr steam output since 1969
	Solvay	Babcock-Atlantique	- 110,000 lb/hr steam output since 1970
	Casablanca	Babcock-Atlantique	- 254,000 lb/hr steam output since 1967
	La Rochette	Babcock-Atlantique	- 110,000 lb/hr steam output since 1961

(Contact: Godel A. A. Societe Anonyme Activit, 66 Rue d'Auteuil, Paris, XVI^e, France)

Referencos:

(1) Godel A. A. d Cosar P. "The Scale-up of a Fluidized-Bed Combustion System to Utility Boilers", AIChE Symposium Seriea, No. 116, Vol. 67, (1971).

(2) Svoboda J. J., "Ignifluid Contribution to Air Pollution Control", The 2nd International Conference on Fluidized-Bed Combustion, Hueston Woods, Ohio, (1968).

7.4. Two-Stage Fluidized Combustion

Description of the Process. This two-stage fluidized combustion concept has been developed in the Fuel Research Institute, Czechoslovakia, since 1952. Combustion of solid fuel with ash content as high as 75 percent without clinker formation is carried out in the first stage fluidized combustor at temperatures below 1000 C. Unburned combustion gas and particles from the first stage are burned in a second combustion space (such as cyclone furnace) at higher temperatures from 1000 to 1200 C. There is no need for immersing heat transfer tubes in the fluidized combustor to recover heat for burning low grade fuels. It is demonstrated that the combustion efficiency improves as the ash content of the fuel increases, a unique feature of this combustion process. It was also demonstrated that the fluidized furnace can burn solid and liquid fuels with 100 percent interchangeability, semioperational and operation boilers, and retrofit of old boilers up to 25 MW were demonstrated.

TABLE 7.4. TWO-STAGE FLUIDIZED COMBUSTION STATE OF THE ART

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Fluidized Furnace lab scale	Prague, Czechoslovakia	Fuel Research Institute	- Research work on the fluidized-bed combustion of solid fuels has been done since 1952 - Fundamental laboratory investigation of small furnaces was conducted in 1958 and 1959 - Two stage combustion of low grade fuel with ash content up to 75 percent, with the first stage at fluidized bed combustion at temperatures less than 1000 C and the second stage (such as cyclone furnaces) at temperatures from 1000 to 1200 C
Semi-operational and operational (Dukafluid)	--	CKD DUKA	- Semioperational fluidized furnaces with capacity of 4 t/hr and 14 t/hr in operation in 1960 and 1961 - Investigation extended to fluid fuels in 1967-1970 and solid and liquid fuels with 100 percent interchangeability was demonstrated.
Retrofit	--	--	- Fluidized furnace retrofitted to an old power plant of 125 t/hr capacity (1971)

(Contact: Pavel Novotný, The Fuel Research Institute, Běchovice, Czechoslovakia)

References:

(1) Pavel Novotný, Sb. Prednášek 50 (Padesatemu) Xýrocí Ústavu Xyzk. Xyužiti, Paliv, 1972.
(2) Pavel Novotný, Technical Digest 1, 1968.

SECTION 8

STACK GAS SCRUBBING

Introduction

The removal of SO_2 from stack gases has been the object of extensive research and development over the past several years. More than 50 individual processes can be identified which are technically feasible in that they have demonstrated ability to remove SO_2 from gas streams. The state of development of these processes varies from bench scale through commercial scale demonstration. Removal processes for SO_2 may be divided into two basic types: throwaway systems in which the SO_2 is converted to a product intended for discard as a solid waste, and recovery systems in which the SO_2 is converted to a useful product. Of the many possible systems, only a few are sufficiently developed for inclusion in this report. They are:

- Throwaway Systems
    - Limestone injection
    - Limestone scrubbing
    - Lime scrubbing
    - Double alkali
- Recovery Processes
    - Wellman-Lord
    - Catalytic oxidation
    - Magnesium oxide
    - Chiyoda
    - Citrate.

Each of these processes is described briefly in the following pages and a summary of the status of operation units is given. Projected start-up dates for units under construction also are included for each type of process.

Status of the Technology

Although the technology for removal of SO_2 from stack gases has been successfully demonstrated for some time, the reduction of the technology to sound engineering practice and widespread acceptance has been slow. This is particularly true from the standpoint of high system reliability which is required for utility application. As experience is accumulated and obvious problems addressed and solved, the availability of the scrubber to the boiler has increased significantly for a number of the test units. Acceptance of SO_2 removal technology is also increasing and the number of units in operation or under construction has increased during the past year. A summary of the installed capacity of units described in this report is given in Table 8.0. There are 15 operational units with a total installed capacity of 2720 MW, with an additional 13 units of 5000 MW capacity under construction. At this stage in the development and application of stack gas scrubbing, there is a heavy dependence on throwaway-type systems. Of the total installed capacity of operational systems, 83 percent is of the throwaway type, while 97 percent of the capacity under construction is throwaway. Since this survey was completed, an EPA report became available entitled, "Flue Gas Desulfurization, Installations and Operations", September 1974. According to that report, the stress on throwaway systems extends to planned units as well. Only 2 of 57 units, for which a process has been selected, are planned to use a recovery-type system.

TABLE 8.0. SUMMARY OF STACK GAS SCRUBBING FACILITIES

Process	MW Capacity (Number of Units)	
	Operational	Under Construction
Limestone injection	785 (4)	
Limestone scrubbing	1128 (4)	2325 (6)
Lime scrubbing	325 (3)	2510 (4)
Double alkali	32 (1)	25 (1)
Wellman-Lord	--	115 (1)
Catalytic oxidation	110 (1)	--
Magnesium oxide	340 (2)	--
Chiyoda	--	25 (1)
Citrate	Pilot Scale	
Totals	2720 (15)	5000 (13)

8.1 Throwaway Processes

8.1.1 Limestone or Lime Injection

Limestone injection involves injection of powdered limestone in the flue gas usually directly in the boiler along with the coal. The limestone then is calcined to lime (CaO) which reacts with SO_2 in the gas stream forming $CaSO_3$ which is removed with a wet scrubber. A schematic of simple limestone injection excluding the wet scrubber is shown in Figure 8.1.1.

The first commercial application of this process was at Union Electric's Meramec Station in St. Louis, Missouri, in 1968. The experiment was terminated in 1971 due to the unusual susceptability of the Meramec boilers to plugging in the convection passages. Other attempts at limestone injection have been made at Kansas Power and Light's Lawrence Plant and Kansas City Power and Light's Hawthorn plant. The Lawrence operation has experienced severe plugging and scaling and may be converted to tail end limestone scrubbing. The Hawthorn plant, after extensive modification, has improved reliability considerably, though sufficient time has not elapsed for realistic evaluation. In one of the two units at Hawthorn limestone is injected downstream of the air heater. The process does operate sufficiently well to permit these two Kansas utilities to meet environmental regulations most of the time.

Limestone injection in general has not been successful enough to warrant plans for future units. In general, the trend is away from limestone injection and towards straight limestone scrubbing.

Lime injection has been tried on an 80 MW unit at the Alma Station of Dairyland Power Cooperative(*) in 1971. Results, however, have not been encouraging as SO_2 removal efficiency is only around 25 percent(**).

(*) Statement of Dairyland Power Cooperative to the State of Wisconsin Department of Natural Resources Public Hearing (July 8, 1972).

(**) Electrical Week, March 10, 1975.

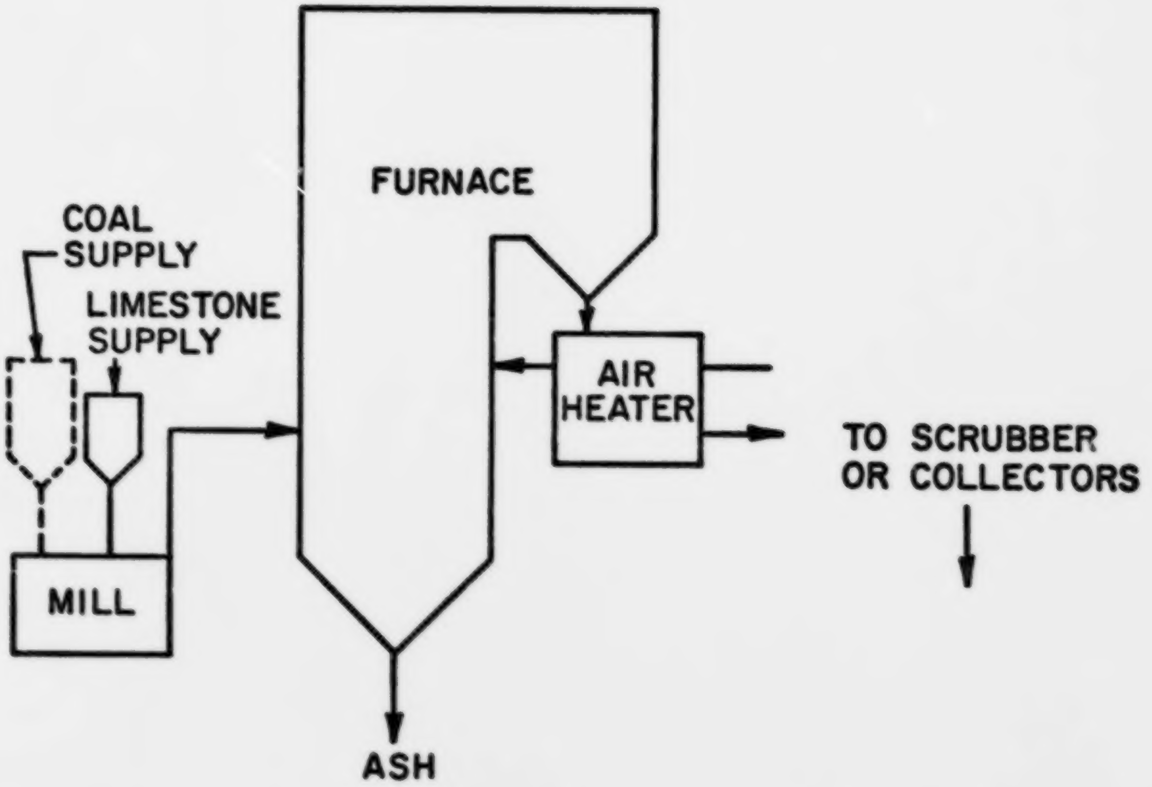


FIGURE 8.1.1. LIMESTONE INJECTION

TABLE 8.1.1. LIMESTONE INJECTION

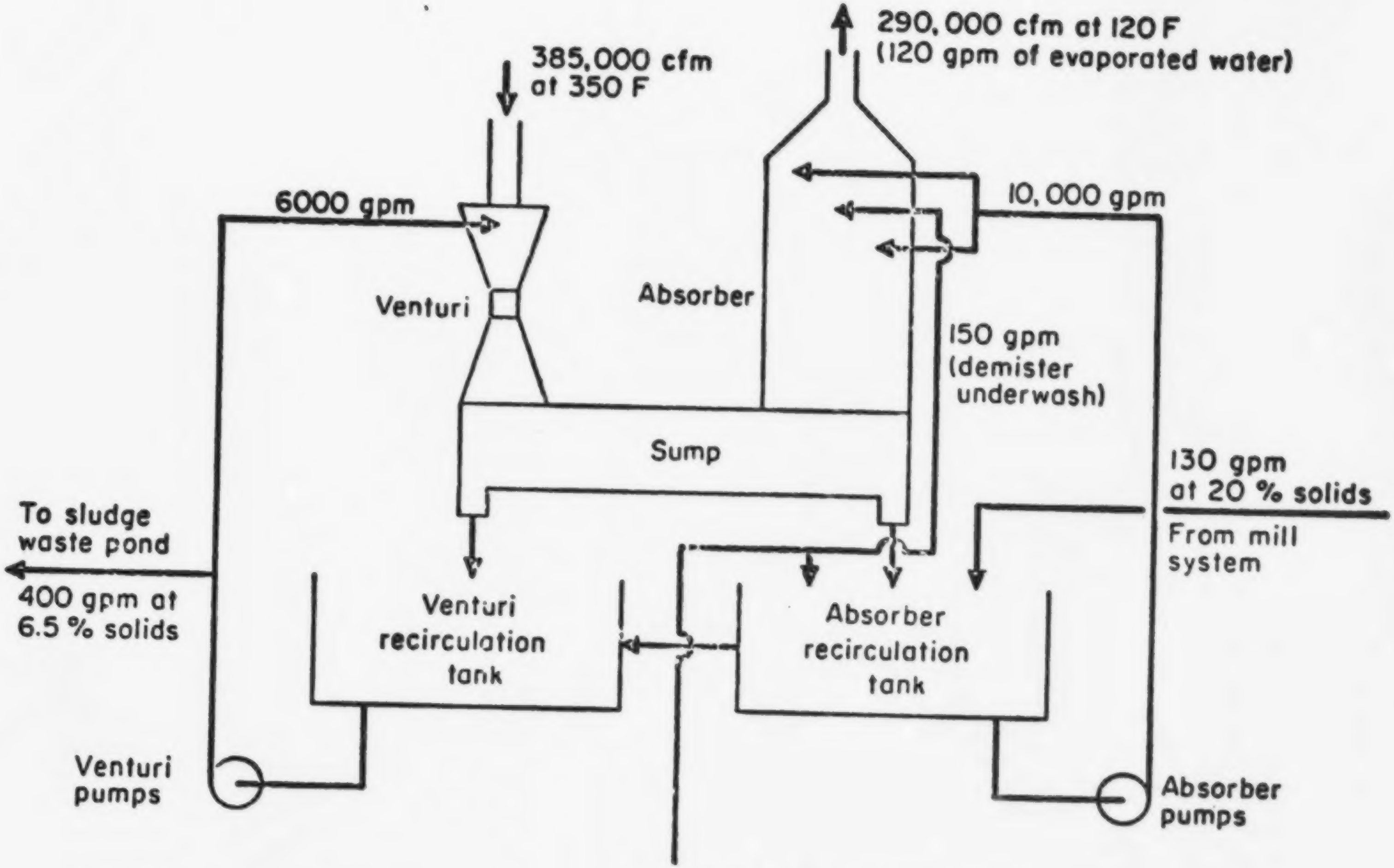
Facilty	Location	Owner(s) or Contractor	Status/Operating History
Meramec 140 MW	St. Louis, Missouri	Union Electric Co., Combustion Engineering	Started up in September 1968. Unit experienced severe plugging problems in the boiler convection passages due in part to the boiler design. Scaling and deposition in ID fans was also a problem. Project was terminated in June 1971.
Lawrence #4 125 MW	Lawrence, Kansas	Kansas Power & Light & Combustion Engineering	Started up in October 1968. Unit has experienced low SO_2 collection efficiency and severe scaling. Half of unit is taken out of service nightly for cleaning. Unit may be converted to straight tailend limestone scrubbing.
Lawrence #5 400 MW	Lawrence, Kansas	Kansas Power & Light & Combustion Engineering	Started up in September 1971. Unit has experienced scaling in the marble bed scrubber and plugging of the demister similar to unit #4. Unit 5 may also be converted to straight tailend limestone scrubbing.
Hawthorn #3 & #4 130 MW	Hawthorn, Kansas	Kansas City Power & Light; Combustion Engineering	Unit #3 started in November 1972, and Unit #4 in August 1972. Both units were originally straight boiler injection of limestone but Unit #4 was converted to inject limestone downstream of the air heater to prevent calcining. Both units initially experienced severe plugging and scaling. After many modifications reliability has been significantly improved, but more time is necessary for a realistic evaluation.

8.1.2 Limestone Scrubbing

Limestone scrubbing is very similar to lime scrubbing except the absorbent is $CaCO_3$ instead of $Ca(OH)_2$. The advantage of limestone scrubbing is that the calcining operation converting limestone into lime can be avoided resulting in lower cost and energy consumption. Figure 8.1.2 gives a simplified process flow sheet for a typical limestone scrubbing installation.

Limestone scrubbing installations, however, have initially shown less success than lime scrubbing installations. The major problems are scaling in the scrubber, plugging of the demister, and corrosion and erosion of stack gas reheat tubes. The problem of scaling has been especially troublesome in closed loop operation where the calcium level reaches saturation and forms scale ($CaSO_4$). In one case, Arizona Public Service's Cholla plant, good operation has been achieved since December 1973 The pond evaporation rate is high and a relatively large fresh water make up rate is permitted minimizing problems with saturation and scaling. On closed loop operation, where the evaporation rate is much less, high reliability has not yet been established although considerable progress is being made at Will County.

At least six new limestone scrubbing installations have been planned, however, making it the most popular form of SO_2 removal in the United States at the present time. Limestone scrubbing is a throwaway process and sludge disposal may be a serious problem in many cases.



Note: Flow rates are for
one module serving about
100 MW of boiler capacity.

FIGURE 8.1.2. LIMESTONE SCRUBBING PROCESS

TABLE 8.1.2. LIMESTONE SCRUBBING PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Will County 156 MW	Joliet, Illinois	Commonwealth Edison; Babcock and Wilcox	Unit started up in February 1972. Major problem has been in sludge disposal (sludge has characteristics of quicksand). Other problems have been in plugging of demisters and corrosion of reheat tubes.
Stock Island 37 MW (Oil)	Key West, Fla.	City of Key West; Zurn Industries	Startup in August 1973. System uses a sea water slurry of native coral as the scrubbing medium. System has had minimal operating experience due to problems in controlling the liquid level in the scrubber. Sludge disposal on the small island is also a problem.
La Cygne 820 MW	La Cygne, Kansas	Kansas City Power & Light; Babcock & Wilcox	Startup in February 1973. Seven module system with only two modules completed thus far. System is similar to that of Will County. Each of the seven scrubber modules is capable of being isolated from the system for repair and full load can be achieved on six modules. Unit has experienced problems with nozzle pluggage. Due to combined problems with scrubber and generating plant little operating data is available.
Cholla 115 MW	Joseph City, Ari.	Arizona Public Service; Research Cottrell	Started up in December 1973. Scrubber has operated well with 80 to 90 percent availability and greater than 90 percent SO_2 removal. Low rainfall and high pond evaporation rates minimize problems of closed loop operation, however. Some problems have been experienced in corrosion of expansion joints and flue gas reheater tubes.
St. Clair 180MW	East China Township, Michigan	Detroit Edison; Peabody	Expected to startup in 1974. Uses Lurgi venturi scrubber followed by tray tower.
Mohave 160 MW	Bullhead City, Nevada	Southern California Gas & Electirc, Universal Oil Prod.	Expected to start up in 1974.
Widows Creek, 550MW	Stevenson, Ala.	TVA	Expected to start up in 1975.
Sherburne County 2 units 680 MW each		Northern States Power; Combustion Engineering	1 unit expected to start up in 1976, the other in 1977.
Gibson 75 MW	Indiana	Public Service of Indiana; Combustion Engineering.	Expected to start up in 1976.

8.1.3 Lime Scrubbing

Lime scrubbing is a wet process for removing SO_2 from flue gas using hydrated lime ($Ca(OH)_2$) as the absorbent. The hydrated lime is usually ground to a fine powder and introduced to a scrubber in a water slurry where it reacts with SO_2 in the flue gas to form $CaSO_3$ and $CaSO_4$. These constituents are then precipitated and settled out of the liquid slurry either in tanks or ponds and disposed of by landfill. A simple schematic diagram is shown in Figure 8.1.3.

Of all flue gas desulfurization processes, lime scrubbing has achieved the greatest success in this country thus far. This is due in large part to the successful operation of Louisville Gas and Electric's Paddy's Run station between April 1973 and December 1973. During this time the unit achieved 90 percent availability with relatively few problems using carbide sludge as the reactant. Carbide sludge is a waste product of acetylene manufacture and is similar to hydrated lime though hydrated lime has not been proven equivalent for scrubbing pruposes. The Paddy's Run station has seen little operation recently due to the boiler heat rate being too high for economical plant operation.

Two other attempts at lime scrubbing in the United States are the Duquesne Light Phillips Station and the Southern California Edison Mohave Station. Phillips has experienced problems with corrosion and erosion including stack leakage and is currently being operated primarily for particulate removal with 50 percent SO_2 removal. Mohave is a recent proprietary installation and only sketchy information is available.

There are currently four new sizable units planned for lime scrubbing. Two are at Bruce Mansfield Station of Pennsylvania Power Company and two at Conesville Station of Columbus and Southern Ohio Edison. Two major problems with lime scrubbing are disposal of the waste sludge (throwaway process) and the expense and energy necessary to make lime from limestone.

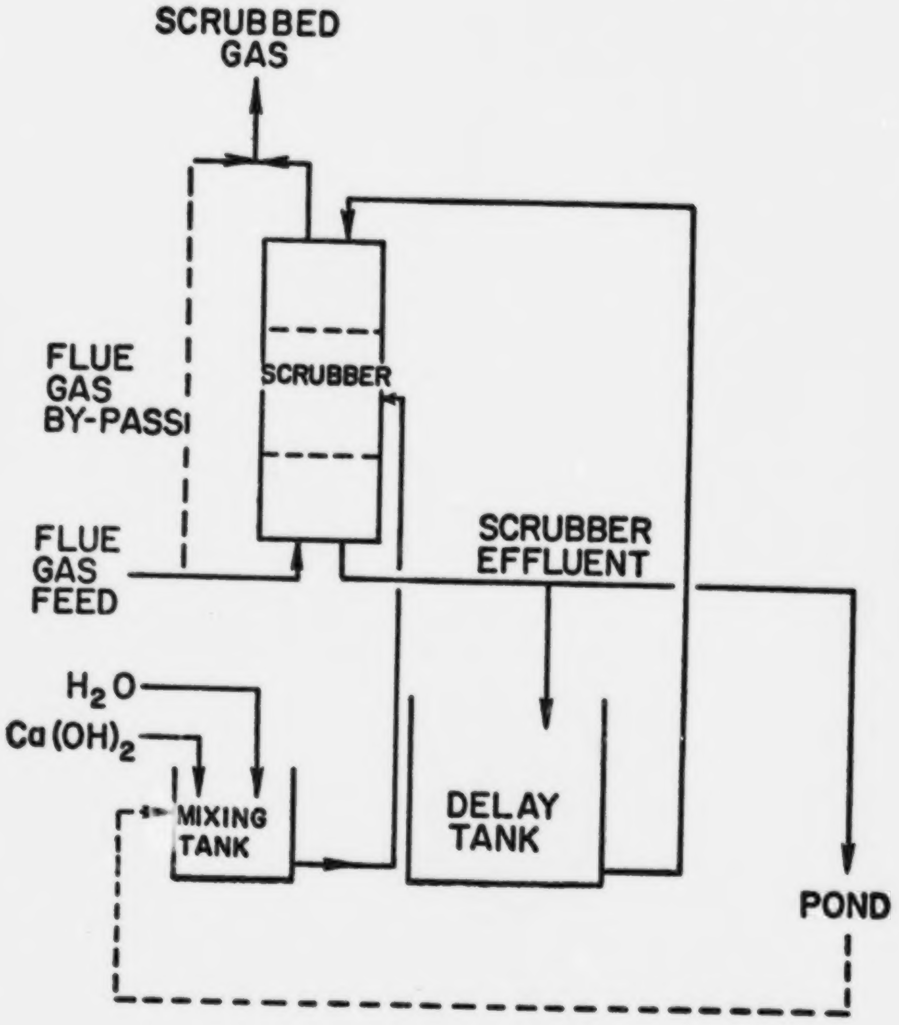


FIGURE 8.1.3. LIME SCRUBBING

TABLE 8.1.3. LIME SCRUBBING

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Paddy's Run 65 MW	Louisville, Ky.	Louisville Gas & Electric; Combustion Engineering	Started up in April 1973. System used carbide lime sludge (similar to hydrated lime $Ca(OH)_2$). Perhaps the most successful SO_2 removal demonstration project in the U.S. to date. From startup to December 1973, system demonstrated 90 percent availability. System was shut down due to high heat rate and uneconomical boiler operation and not because of scrubber problems. System used a borrow pit for sludge disposal and natur natural gas for stack gas reheat.
Phillips 180 MW unit 100 MW scrubber	So. Heights, Pa.	Duquesne Light; Chemico	Started up in April 1973. First demonstration of use of hydrated lime ($Ca(OH)_2$) in the U.S. System was shut down in October 1973 due to leakage in the stack, corrosion in ID fans and corrosion and erosion in scrubber. System was restarted for flyash collection with reactant addition sufficient to prevent corrosion and give over 50 percent SO_2 removal.
Mohave 790 MW unit 160 MW scrubber	Bullhead City, Nevada	Southern California Edison; Stearns-Roger	Started up in January 1974. Scrubber is a unique horizontal type with an electrostatic precipitator upstream for flyash removal. Availability has been over 80 percent. Major problems have been in corrosion and erosion of pumps. Unit has not used high sulfur coal (coal sulfur is around 0.5 percent) and is in an area where high pond evaporation occurs minimizing problems of closed loop operation.
Bruce Mansfield Units #1 & #2 Both units 880 MW	Shippingport, Pa.	Pennsylvania Power Co. Chemico	Unit #1 to start up in 1975. Unit #2 to start up in 1976.
Conesville 2 units 375 MW each	Conesville, Ohio	Columbus & Southern Ohio Electric Co. Universal Oil Products.	Units to start up in 1976.

8.1.4 Double Alkali

The double alkali processes involves the use of NaOH or an ammonia salt as a primary absorbent for removing SO_2 from the flue gas followed by reaction with lime or limestone to regenerate the primary absorbent and yield an insoluble waste product ($CaSO_4$). A simplified schematic diagram of the process is shown in Figure 8.1.4.

One of the main problems with the double alkali process is the difficulty in regenerating Na_2SO_4. The industrial boiler of the General Motors Parma plant is currently the only commercial double alkali process in operation and has experienced some difficulty with plugging in the causticizer, but operation is improving with experience.

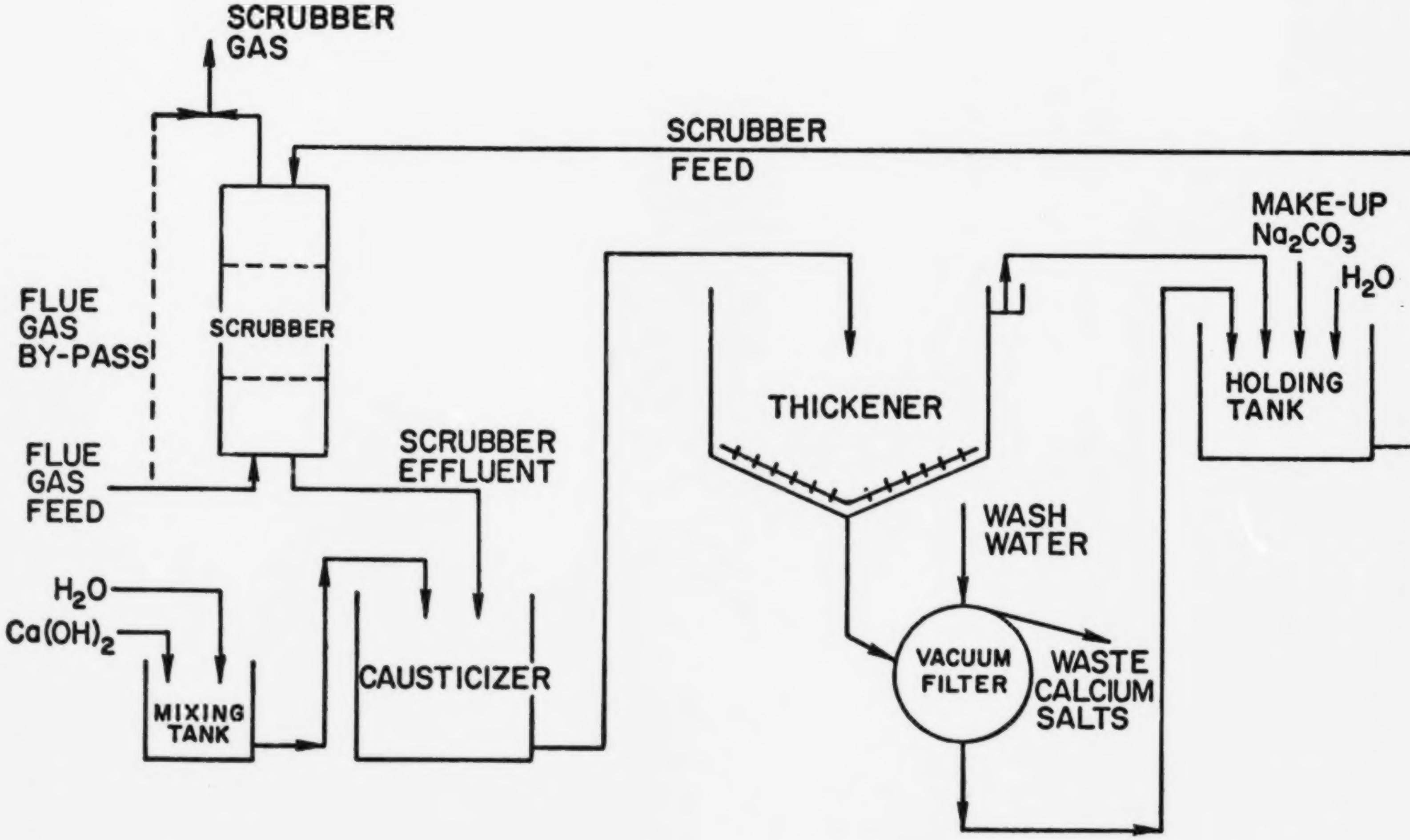


FIGURE 8.1.4. SODIUM SCRUBBING WITH LIME REGENERATION (Double Alkali)

TABLE 8.1.4. DOUBLE ALKALI (SODIUM SCRUBBING-LIME REGENERATION)

Facilty	Location	Owner(s) or Contractor	Status/Operating History
General Motors 32 MW (steam only)	Parma, Ohio	Argonaut Engr., Div. of GMC; Combustion Equipment Assoc.	Started up in April 1974. The first major problem has been caking in the causticizer (see Figure 8.1.4). Insufficient time has elapsed for effective evaluation.
Caterpillar Tractor 2 units 10 MW } steam only 8 MW }	Joliet, Illinois	Caterpillar Corp; Zurn Industries	Units due to start up in 1975.
Caterpillar Tractor 15 MW } steam only 8 MW } 8 MW }	Mossville, Ill.	Caterpillar; Food Machinery Company	Unit due to start up in 1975
Gadsby Station 2500 cfm pilot	Salt Lake City, Utah	Utah Power & Light; Envirotech Corp.	Unit started up in early 1974. Uses NaOH to scrub flue gas followed by CaO to regenerate the absorbent and yield an insoluble waste product. Envirotech is offering system for full scale application.
Scholz 25 MW	Sneads, Florida	Gulf Power Combustion Equipment Associates	Anticipated startup is in 1975.

8.2 Recovery Processes

8.2.1 Wellman Lord Process

The Wellman Lord Flue Gas desulfurization process is a wet process using a concentrated caustic solution (sodium sulfite) as the absorbent. In removing SO_2 from the gas stream sodium sulfite is converted to sodium bisulfite which is then disproportionated with heat (steam) to regenerate sodium sulfite while forming a concentrated SO_2 gas (90 percent SO_2). The sodium sulfite is returned to the absorber and the concentrated SO_2 stream can be processed to sulfuric acid or elemental sulfur by conventional means. SO_2 removal efficiency of the process is believed to be greater than 90 percent. The process is handled by Davy Powergas of Lakeland, Florida. A schematic flow diagram of the process is shown in Figure 8.2.1.

The first boiler application of the Wellman Lord process was on the Japan Synthetic Rubber Chiba plant in 1971. The facility serves two oil fired boilers with a capacity of 286,000 pounds of steam per hour each. The first U.S. installation on flue gas, and first known installation on coal will be the Northern Indiana Public Service Company's Mitchell Station at Gary, Indiana.

There is concern that impurities in the coal ash could accelerate the undesired formation of sodium sulfate when sodium sulfite is oxidized. Sodium sulfate along with other impurities must be purged from the scrubbing liquor with a bleed stream and represent a disposal problem. At Chiba about 10 percent of the sodium content is lost on each pass through the scrubber as a result of this purge. Flyash in the regeneration loop also must be purged.

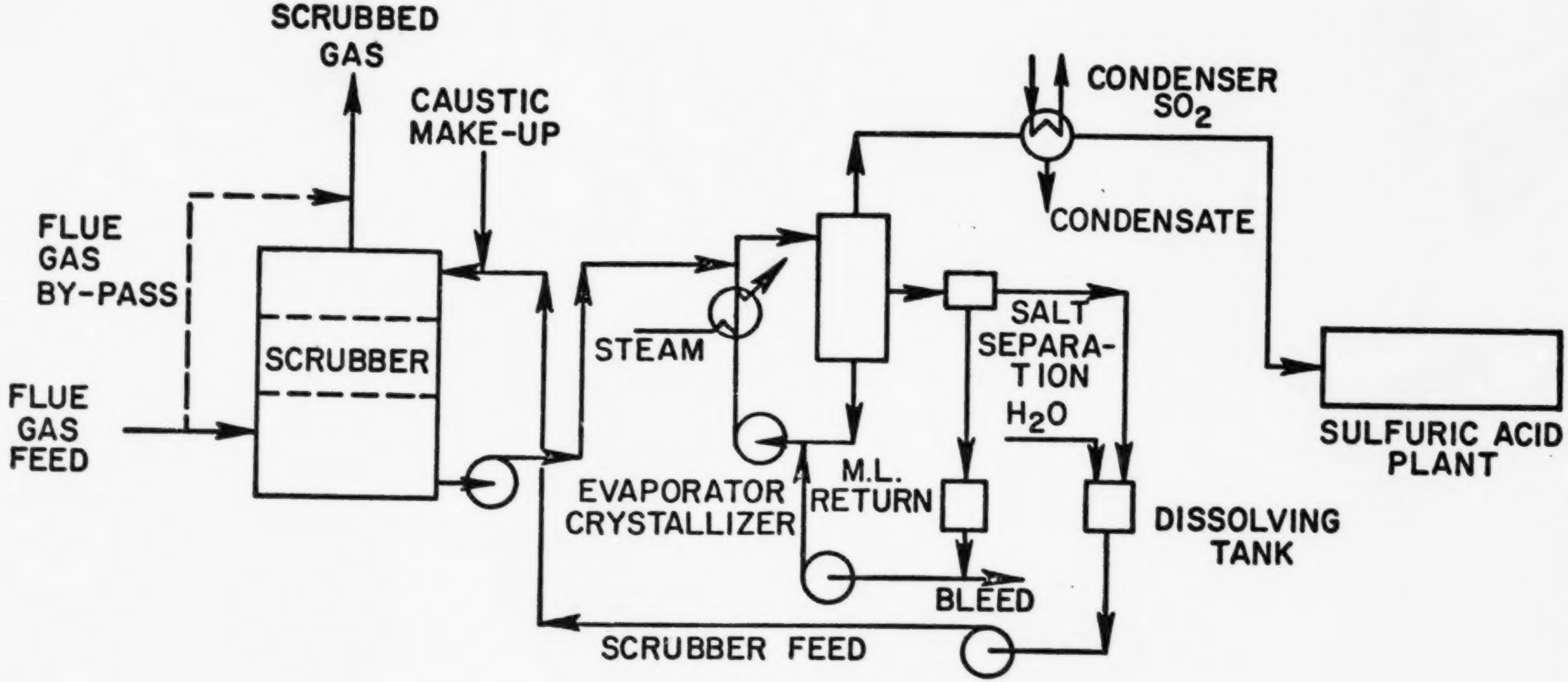


FIGURE 8.2.1. SODIUM SCRUBBING WITH SULFUR DIOXIDE RECOVERY (Wellman Lord)

TABLE 8.2.1. WELLMAN LORD PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Mitchell Station	Gary, Indiana	Northern Indiana Public Service Co. & Environmental Protection Agency	Will be the first boiler application of process in U.S. Process will be applied to a 115 MW coal fired utility boiler. The recovered concentrated SO_2 stream will be reduced with natural gas to elemental sulfur (Allied Chemical Process). Process has been used successfully on oil fired boilers in Japan and on SO_2 tail gas from Claus units in refineries.

8.2.2 Catalytic Oxidation

The catalytic oxidation process (CAT-OX) uses a fixed bed catalytic converter to oxidize the SO_2 in the flue gas to SO_3 which can then be collected in an absorption tower with water resulting in about 78 percent sulfuric acid. The flue gas must be at about 850 F entering the converter. For a new power plant installation, this could be done directly. In a retrofit application, flue gas temperatures are from 250 F to 350 F leaving the air preheater thus requiring the flue gas to be heated to 850 F with an auxiliary fuel (oil or natural gas). Much of the heat put in is recovered in a regenerative heat exchanger. A simplified schematic diagram is shown in Figure 8.2.2.

There is only one commercial scale application of the CAT-OX process; a 110 MW coal fired boiler at the Wood River Station of Illinois Power. The concept has been demonstrated briefly and sulfuric acid produced. The major problems have been in obtaining suitable gas reheat. Originally the system was designed for firing natural gas directly into the flue gas for reheat, but, shortages have forced the use of oil. There have been problems with temperature control with oil and also the possibility of oil soot contaminating the catalyst. A one-year test program will start in early 1975, during which time it is hoped that feasibility will be demonstrated.

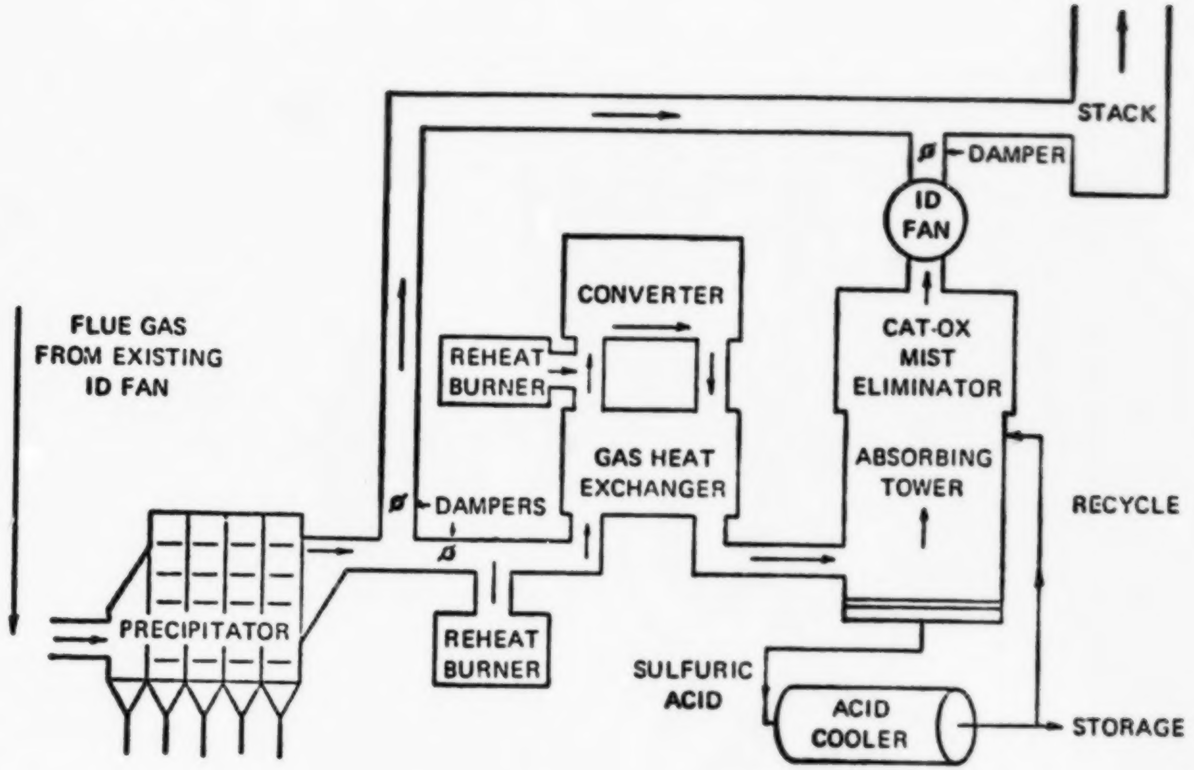


FIGURE 8.2.2. CATALYTIC OXIDATION

TABLE 8.2.2. CATALYTIC OXIDATION

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Wood River 110 MW (coal)	Wood River, Ill.	Illinois Power; Monsanto	System originally started up in 1972. Major problems have been in controlling gas reheat temperature at catalytic converter inlet using fuel oil. The effect of flyash on catalyst life also has not yet been determined.

8.2.3 Magnesium Oxide Scrubbing

The MgO wet desulfurization process utilizes finely ground magnesium oxide in a water slurry as the scrubbing medium in conventional scrubbing equipment. The MgO reacts with the SO_2 in the flue gas forming $MgSO_3$ which is dewatered and sent to a sulfuric acid plant to regenerate the MgO. Regeneration is accomplished by heating the $MgSO_3$ in a rotary kiln yielding MgO and an SO_2-rich gas. The SO_2-rich gas is then processed through a conventional contact sulfuric acid plant to make a commercial grade sulfuric acid. A simplified schematic flow sheet is shown in Figure 8.2.3.

In the two full-scale demonstration projects of MgO scrubbing in this country the scrubbing process itself has shown promise. The major problems in both cases have been mechanical failures of piping, pumps, and related equipment. Also,some doubt still exists as to the ability of the unit to operate over a long term on regenerated MgO with a minimum of fresh MgO makeup. This is an important consideration due to the high cost of magnesium oxide. Also, MgO units operating on coal will require a purge stream to control the flyash concentration in the regeneration loop.

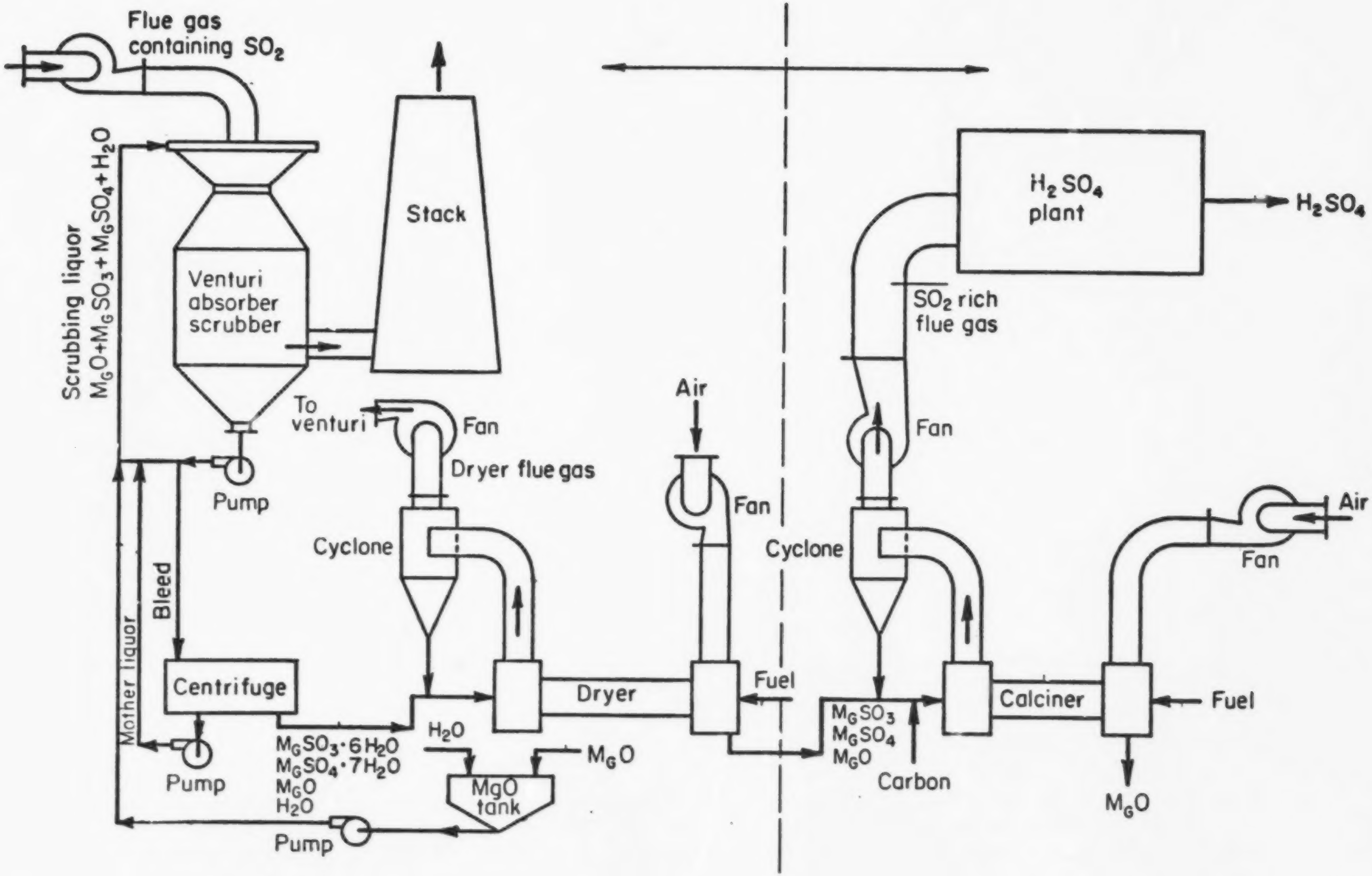


FIGURE 8.2.3. MAGNESIUM OXIDE SCRUBBING WITH SULFURIC ACID RECOVERY

TABLE 8.2.3. MgO SCRUBBING

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Mystic 150 MW (oil)	Boston, Mass.	Boston Edison; Chemico	Unit was first started up in April 1972. Unit was initially plagued with mechanical problems such as corrosion and erosion of pumps valves and piping. Through proper equipment selection and experience, progress has been made in this area. SO_2 removal has been over 90 percent when the scrubber is operating. Scrubber availability has averaged less than 50 percent over its operating life. No decision has been made to equip other units.
Dickerson 190 MW	Dickerson, Maryland	Potomac Electric Power; Chemico	Unit started up in 1974. Major problems have been in mechanical failures external to the scrubber. MgO scrubbing process has worked satisfactorily.

8.2.4 Chiyoda Process

The Chiyoda "Thoroughbred 101" flue gas desulfurization process is a wet process which removes SO_2 and SO_3 from a gas stream by absorption in dilute H_2SO_4. After oxidation of the $SO_3^=$ to sulfate ($SO_4^=$) with the aid of a soluble catalyst, $Fe_2(SO_4)_3$, part of the H_2SO_4 is reacted with a calcium compound such as limestone to form gypsum. The remainder of the H_2SO_4 is recycled to the absorber as shown in the schematic flow diagram in Figure 8.2.4.

This process was originally designed for oil fired systems and in order to use it on coal fired systems some modifications are necessary.

(1) An efficient particulate collection device is needed ahead of the absorber to reduce the dust concentration to an acceptable level. Since most coal fired utility boilers in this country already have electrostatic precipitators for fly ash collection this may not be a problem, although, collection efficiency may have to be increased in some cases.

(2) A vacuum filter may be needed for removing the residual fly ash from the dilute H_2SO_4 to prevent flyash contamination of the gypsum.

This process has never been reportedly applied to a coal fired boiler, although there are five installations on oil fired boilers in Japan. Chiyoda recently announced plans to install a 23 MW demonstration plant at the coal fired Scholz plant of Gulf Power Company near Sneads, Florida as the first attempt. Even with efficient flyash collection enough dust may penetrate to the absorber and cause plugging. The ash may also cause contamination of the gypsum and impair its value as a salable product.

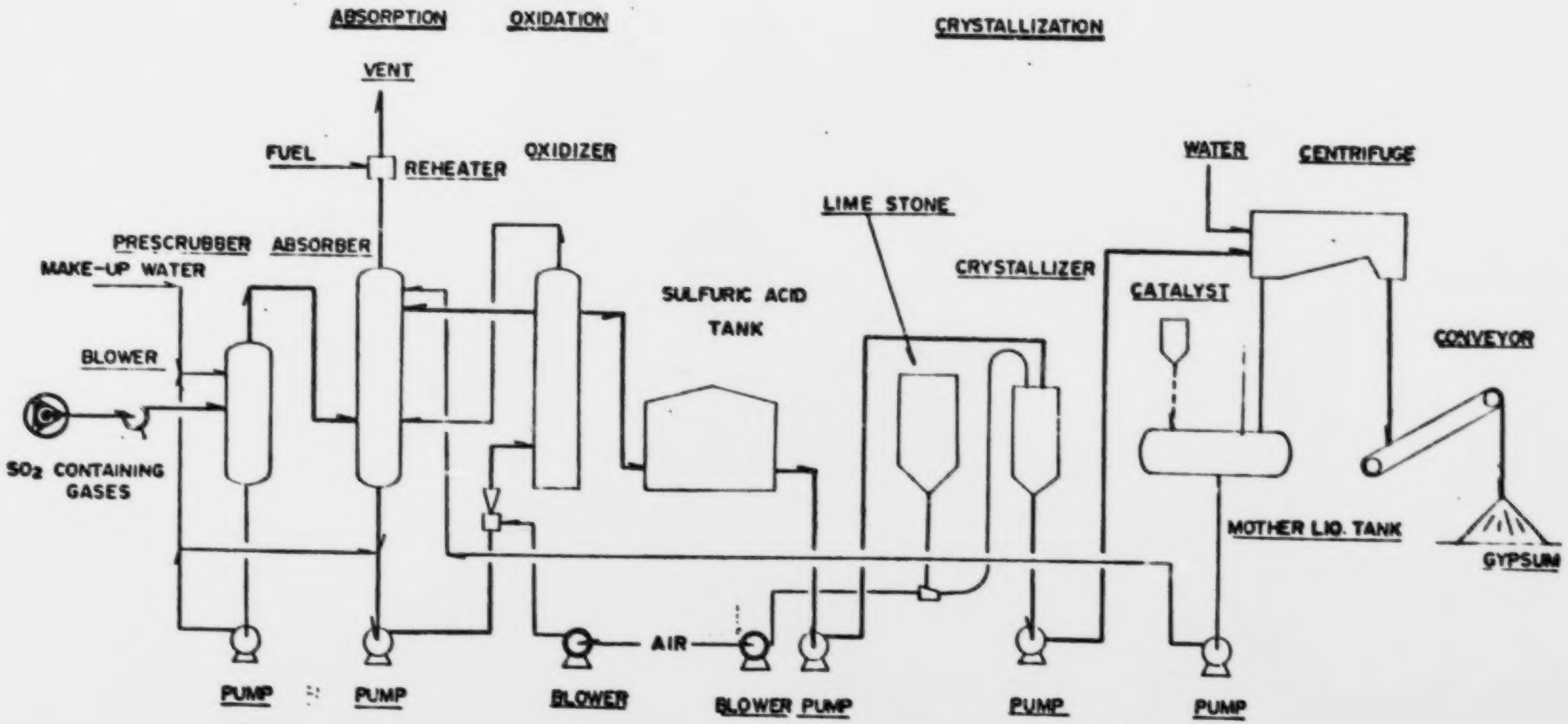


FIGURE 8.2.4. CHIYODA THOROUGHBRED 101 FLUE GAS DESULFURIZATION PROCESS

TABLE 8.2.4. CHIYODA PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Scholz 25 MW	Snead, Florida	Chiyoda Chemical Engineering and Construction Co.	Plans were announced in December 1973 to build a 23 MW demonstration unit. Will be the first Chiyoda unit in the U.S. and the first anywhere on coal. There are five Chiyoda units on oil fired installations in Japan.

8.2.5 The Citrate Process

The Citrate process involves absorption of the SO_2 in the flue gas in citric acid ($C_6H_8O_7$) or some other carboxylate solution followed by reaction of the spent solution with H_2S in a closed vessel to precipitate the absorbed SO_2 as elemental sulfur. First the flue gas must be washed to remove particulates and SO_3 before entering the absorber. The sulfur precipitate is removed from the regenerated solution by oil floatation and melting. A schematic flow diagram of the Citrate process is shown in Figure 8.2.5.

This process was developed by the Bureau of Mines at Salt Lake City and tried on a small scale at the San Manuel copper smelter near Tucson, Arizona beginning in November 1970. The pilot plant (300 cfm) was plagued with failures of the gas cleaning system, pump breakdowns, and plugging of flow lines with precipitated and melted sulfur. SO_2 removal of 90 to 99 percent was readily obtainable.

The process is now being developed by Pfizer with support from the Bureau of Mines, Peabody Engineering Co. and Arthur F. McKee. A 2000 cfm pilot plant, built on skids, is now operating at a 150 MW power plant in Terre Haute, Indiana on 3.5 percent sulfur coal. The chemical process was generally been satisfactory but the unit has been troubled with mechanical failures. Thus far methane (natural gas) has been used to generate H_2S from elemental sulfur.

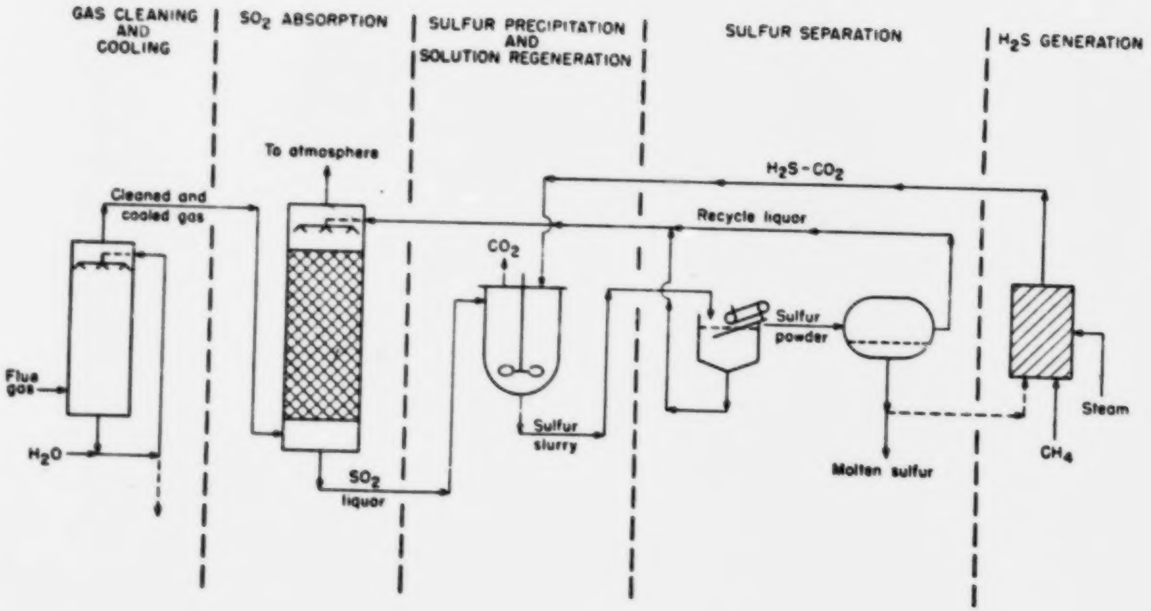


FIGURE 8.2.5. THE CITRATE PROCESS

TABLE 8.2.5. CITRATE PROCESS

Facilty	Location	Owner(s) or Contractor	Status/Operating History
Terre Haute 2000 cfm	Terre Haute, Ind.	Pfizer Chemical; Peabody Engineering; Arthur F. McKee	Pilot plant unit mounted on skids. Has had mechanical failures though chemical process has proven satisfactory. Much information is proprietary at this time.

Selected Bibliography for Section 8

(1) Oxley, J. H. et al, Sulfur Emission Control for Industrial Boilers, paper presented at the American Power Confernece (April 1974).

(2) Land, G. W., Nichoson, F. E., Status of Flue Gas Desulfurization Projects, Amax Coal Co. (May 1974).

(3) Rosenbaum, J. B. et al, Sulfur Dioxide Emission Control by Hydrogen Sulfide Reaction in Aqueous Solution, Bureau of Mines (1973).

(4) Rosenberg, H. S. et al, State of the Art Report on Status of Development of Process for Abatement of SO_2 Emissions by Stack Gas Treatment, report to American Electric Power Service Corporation (March 1973).

(5) Flue Gas Desulfurization, Installations and Operations, U.S. Environmental Protection Agency (September 1974).

(6) National Public Hearings on Power Plant Compliance with Sulfur Oxide Air Pollution Regulations, U.S. Environmental Protection Agency (January 1974).

SECTION 9

ACKNOWLEDGEMENTS

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APPENDIX A

ENVIRONMENTAL CONSIDERATIONS FOR THE GASIFICATION OF COAL

Gasification of coal has associated with it a number of potentially undesirable environmental effects which must be held within acceptable limits. Toward that end, EPA has initiated a continuing program aimed at providing environmental assessments and the necessary emissions control and abatement technology for synthetic fuel processes. This program will operate in parallel with the gasification research and development programs described earlier (Sections 5 and 6). Hopefully, potential environmental problems will be characterized well enough and soon enough to permit proper abatement systems to be incorporated into the design and construction of commercial plants.

Applications have already been filed with the Federal Power Commission for the construction of two commercial-scale (250×10^6 SCF/D) plants for the production of SNG using the well-developed Lurgi process (see Table 6.1). The possible environmental impact of these plants has received careful attention from the designers and exacting scrutiny by the various regulatory agencies. Operating experience at these installations should provide valuable feed back.

The environmental impact of coal gasification processes has received considerable attention (3, 6, 7, 28, 29, 30, 31)*; and for purposes of this report a brief outline of the major problem areas should be sufficient.

Coal Mining. Although the environmental problems will be of the same type as presently encountered in coal production, the magnitude of the mining effort that will be required to support a mature coal-gasification industry is such that this aspect warrants very careful attention.

The two commercial-scale plants to be constructed in New Mexico will each require about 25,000 tons of coal per day to produce 250×10^6 CFD of SNG. By 1990 it is estimated that SNG production alone will amount to 1.6 x 10^{12} ft^3/yr*. This will require about 100-150 x 10^6 ton/yr of coal**; in constrast, total U.S. production was about 613 x 10^6 ton/yr in 1970 (33).

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*References cited in this Appendix refer to items contained in the Supplemental Bibliography for Coal Gasification on Page 6-37.

Also, because the cost of coal is an important factor in determining the price of the product gas***, there will be a strong economic incentive to use surface-mined coal whenever possible. This will present a particularly challenging problem in the arid regions of the West where land reclamation is difficult.

At present, there are no Federal Regulations governing restoration of land following surface mining. However, the Senate and House both have recently passed relatively strict laws, and a conference committee is now meeting to resolve differences in the two bills. The overwhelming vote in the House (291-81) suggests that there is sufficient support to override a possible veto.

Coal Preparation. Coal cleaning, drying and sizing can result in air and water pollution as well as a solid waste problem. However, as in the case of mining, the problems are no different than those now associated with the production of coal. The specific pollutants and the processes by which they reach the environment are reasonably well understood; and control equipment is commercially available.

The solid wastes produced during coal cleaning may require a significant commitment of land resources. A modern 1,000 TPH cleaning plant requires between 40 and 90 acres for waste disposal (6).

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* "The Coal Gasification Market", prepared by marketing consultants Frost & Sullivan of N.Y. A brief summary of some of their major findings appears in Chemical Engineering, pp 59-60 (July 22, 1974). This figure is in reasonable agreement with estimates in the National Petroleum Councils, U.S. Energy Outlook: An Initial Appraisal, 1971-85 (November 1971). See also Reference 6.

** Based on data in Reference 33, pp 58-70.

*** For example, a $1.00 increase in the cost of coal results in a 13¢/MMBtu increase in the cost of SNG produced by the Lurgi process (6).

Coal Gasification. A number of products are recovered in various process streams which are potential sources of pollution. The variety and nature of these products depend upon operating conditions and the basic unit operations involved (3, 6, 28, 30, 32). Some of these products such as tar, oils and char can be recycled and others such as phenols, ammonia and sulfur may be marketable byproducts. Table A.1 indicates the yields of products expected from a 250 MMCFD gasification plant operating on Illinois No. 6 coal (3.7 percent S). Ranges are necessary because of dependence of yields upon the nature of the gasification process.

A. J. Forney, et al., carried out extensive studies of the various effluents associated with the Synthane coal-to-gas process (32), and they have published the analyses of tars, chars, gases and waters produced as effluents when different coals are used. Although yields can vary quantitatively and qualitatively for different processes, the results are indicative of the complexity of these streams.

Large quantities of water are required in the gasification of coal. For example, the 250 MMSCF/D WESCO plant will require 5,100 gpm of raw water intake (8,200 acre-feet per year). This amounts to about 1.4 pounds of raw water intake per pound of coal. In general, the higher the heating value of the product gas, the greater the water requirements per unit of product gas. Some large western coal deposits which are likely to be considered for gasification are in water-deficient areas. This may not be a serious constraint in the early stages of development, but the overall management of water resources in these areas should be an important consideration.

In addition to serving as a raw material in gasification i.e, as a source of hydrogen), water is also involved in a number of process steps such as cooling and scrubbing*. The studies by Forney et. al. on the chemical composition of aqueous process streams indicates their complexity (Tables A.2 and A.3). In

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(5) In the Lurgi process being used at WESCO about 10 percent of the water will be consumed in gasification, 70 percent will be returned to the regional atmosphere and the remaining 20 percent is disposed of on-site -- principally as sludge and wetted ash. There will be no return of waste water to the source (San Juan River). (Based on information in "Coal Gasification: A Technical Description, published by WESCO.)

TABLE A.1. BYPRODUCT YIELDS IN COAL GASIFICATION(a)

Product	Yield (long tons/day)
Sulfur	300-450
Ammonia	100-150
Hydrogen Cyanide	0 to possibly 2
Phenols	10-70
Benzene	50-300
Oil and Tars	Trace to 400
Mercury	Less than 5 lbs/day

(a) For production of 250 MMCFD of SNG from Illinois No. 6 Coal (3.7%S). Taken from Reference 3 page X-2.

TABLE A.2. BYPRODUCT WATER ANALYSIS FROM SYNTHANE GASIFICATION OF VARIOUS COALS, MG/1 (EXCEPT pH)(32)

	Coke plant	Illinois No. 6 coal	Wyoming subbi-tumi-nous coal	Illi-nois char	North Dakota lignite	Western Kentucky coal	Pitts-burgh seam coal
pH	9	8.6	8.7	7.9	9.2	8.9	9.3
Suspended solids	50	600	140	24	64	55	23
Phenol	2,000	2,600	6,000	200	6,600	3,700	1,700
COD	7,000	15,000	43,000	1,700	38,000	19,000	19,000
Thiocyanate	1,000	152	23	21	22	200	188
Cyanide	100	0.6	0.23	0.1	0.1	0.5	0.6
NH_3	5,000	[1] 8,100	9,520	2,500	7,200	10,000	11,000
Chloride	-	500	-	31	-	-	-
Carbonate	-	[2] 6,000	-	-	-	-	-
Bicarbonate	-	[2] 11,000	-	-	-	-	-
Total sulfur	-	[3] 1,400	-	-	-	-	-

[1] 85 percent free NH_3.

[2] Not from same analysis.

[3] $S^{=}$ = 400
$SO_3^{=}$ = 300
$SO_4^{=}$ = 1,400
$S_2O_3^{=}$ = 1,000

TABLE A.3. TRACE ELEMENTS IN CONDENSATE FROM AN ILLINOIS NO. 6 COAL GASIFICATION TEST(32)

	No. 1	No. 2	Average (by weight)
ppm			
Calcium	4.4	3.6	4
Iron	2.6	2.9	3
Magnesium	1.5	1.8	2
Aluminum	0.8	0.7	0.8
ppm			
Selenium	401	323	360
Potassium	117	204	160
Barium	109	155	130
Phosphorus	82	92	90
Zinc	44	83	60
Manganese	36	38	40
Germanium	32	61	40
Arsenic	44	28	30
Nickel	23	34	30
Strontium	33	24	30
Tin	25	26	20
Copper	16	20	20
Columbium	7	5	6
Chromium	4	8	6
Vanadium	4	2	3
Cobalt	1	2	2

commercial plants it will probably be economically imperative to purify water for reuse, and therefore there should be no major waste-water effluent except that used to slurry ash for disposal.

Removal of hydrogen sulfide from the raw gas leaving the gasifier is an important consideration. In almost all cases the sulfur is ultimately recovered as elemental sulfur. The various processes involved and their application to coal gasification have been described in detail elsewhere (8,38).

Large quantities of solid waste are produced during gasification[*]. Most of this is ash, but processing to remove sulfur may also contribute to the solid waste burden. Some processes, such as Hygas, CO_2 Acceptor and those involving molten salt baths, generate additional solid waste streams.

In conclusion, gasification does involve potentially undesirable environmental effects. It would be unrealistic to think that these adverse effects can be completely eliminated; however, careful planning and good engineering practice should make it possible to hold them to an acceptable level. Coal mining and preparation are probably the areas that present the most significant environmental problems; this is undoubtedly one of the reasons for renewed interest in underground gasification. However, the technical and environmental feasibility of underground gasification on a large-scale have not yet been demonstrated. Moreover, the apparent attractiveness from an environmental viewpoint may result primarily from the fact that much less is known about the potential environment consequences of underground gasification on a large-scale.

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[*] A typical 250 x 10^6 SCF/day gasification plant will produce between 1,000 and 3,000 ton/day of ash. The CO_2 Acceptor Process will also discharge about 900 ton/day of spent dolomite.